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EVALUATION OF RADIOISOTOPE TRACER AND ACTIVATION  
ANALYSIS TECHNIQUES FOR CONTAMINATION  
MONITORING IN SPACE ENVIRONMENT SIMULATION CHAMBERS

Final Report

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## FOREWORD

This is a final report covering work performed under Contract Number NAS 9-12586 during the period March 15, 1972 to April 1, 1973.

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## ABSTRACT

Radioisotope measurement techniques and neutron activation analysis are evaluated for use in identifying and locating contamination sources in space environment simulation chambers. Methods considered include radioactive tagging of contaminants, charged particle range variation and stable isotope tagging. Of the several methods studied, only the alpha particle range technique offers mass measurement sensitivity capable of competing with the quartz microbalance presently in use. Although unable to identify specific contaminants, the alpha range method allows the determination of total contaminant concentration in vapor state and condensate state with calculated sensitivities of  $1.9 \times 10^{-13}$  g/cm<sup>3</sup> to  $1.9 \times 10^{-10}$  g/cm<sup>2</sup> respectively. These values are applicable to clean dry air or to chamber air contaminated in any manner. A Cf-252 neutron activation analysis system for detecting oils and greases tagged with stable elements is described. While neutron activation analysis of tagged contaminants offers specificity, an on-site system would be extremely costly to implement (\$520,000 plus) and would provide only marginal detection sensitivity under even the most favorable conditions. Off-site reactor neutron activation analysis of tagged samples would provide adequate measurement sensitivity at moderate cost but difficulties associated with sample retention during chamber repressurization would seriously limit the effectiveness of this approach. Also, a delay of from one to two days would be required between sample acquisition and availability of analytical results.

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## 1.0 INTRODUCTION

The objective of this study was to determine the feasibility of several nuclear analytical techniques for the identification and measurement of very small quantities of contaminants in space environment simulation chambers at NASA Manned Spacecraft Center. This project was carried out in two phases; Phase I being directed toward the use of radioactive isotope techniques while Phase II was concerned with the use of activation analysis. The merits of a given technique are evaluated on the basis of measurement sensitivity, ability to identify specific contaminants, speed of analysis, suitability for real-time operation either in-chamber or on-site near the chamber, radiological safety and cost of implementation. In Phase I, many potential methods were initially considered without regards for their radiological safety acceptability. Those which indicated an acceptable sensitivity capability were then evaluated for radiological safety and design feasibility. Section 2.0 of this report covers studies of radioisotope techniques while Section 3.0 reports results of the activation analysis studies.



## 2.0 RADIOISOTOPE TECHNIQUES

### 2.1 SUMMARY OF PRESENT METHODS SENSITIVITIES

A consensus of the reliable sensitivities for some of the various devices currently in use were not readily available. From information on tests run (Apollo Telescope Mount series), a lower detection limit for the Contamination Control Units (CCU) system would appear to be  $1 \times 10^{-9} \text{ g/cm}^2$  (Ref. B-1), while average contamination levels during these tests were on the order of  $1 \times 10^{-7} \text{ g/cm}^2$  (Ref. J-1). The Quartz Crystal Microbalance (QCM) System was quoted as having a minimum detectable limit of  $10^{-9}$  grams (Ref. A-5). It was decided that any system developed must see at least this level and preferably, have a sensitivity less than  $10^{-9} \text{ g/cm}^2$ .

### 2.2 TAGGING OF OILS WITH CARBON-14

For purposes of this calculation it was assumed that  $7 \times 10^{-8} \%$  (Ref. C-14) of the oil molecules could be tagged with one Carbon-14 atom through an isotope exchange process. The oil so tagged would then be used in its normal function and the atmosphere of the chamber monitored for any increase in background activity. Of the detection systems considered,

high pressure gas detectors, scintillation detectors, and solid state detectors; two specific detectors were chosen for incorporation into the study because of their sensitivity to low energy beta emitters and suitability with regard to chamber environmental conditions:

- A. An avalanche detector, placed one meter from a one-meter diameter  $\text{LN}_2$  cooled plate.
- B. A one-meter diameter, radiantly-cooled, solid plastic scintillator.

The sensitivity determination for each type of detector is as follows:

A. Avalanche Detector Analysis

A windowless and an aluminized mylar windowed detector were considered. Assuming that 100 counts per minute above background are necessary for detection and that through suitable pulse height discrimination the background can be made less than 50 cpm, then the necessary activity of the deposited oil on the plate is:

$A = 100/(G \cdot E)$ , where E is the efficiency of the detector and G is the geometry factor of the system.

$G = 0.5 (1 - \cos \theta) = \sin^2(\theta/2)$  (Ref. C-10), where  $\theta$  is the solid angle between detector and source,  $\theta = 26.5^\circ$ , thus  $G = 0.0528$ .

The efficiency of the detector is dependent on the number of particles penetrating both the detector dead layer and the window. Carbon-14 has a 156 keV beta particle and one can use the range energy relationship to estimate the percentage of particles reaching the detector-sensitive volume. Assuming an average 5 micron dead layer and a 0.00025 inch aluminized mylar window, the resulting efficiencies are: with the window,  $E = .72$  and without the window,  $E = .97$ . Assumptions made in the analysis

were a negligible dead time correction, absence of radiation particles scattered by nearby structural materials, and negligible absorption by the residual gas atmosphere in the chamber over the one meter distance. This latter assumption would be invalid during the initial pump down and final warmup stages.

Therefore, with window,  $A = 2.63 \times 10^3$  dpm and without window,  $A = 1.95 \times 10^3$  dpm.

To obtain the number of detectable atoms of Carbon-14, one uses  $N = A/\lambda$ , where  $\lambda$ ,  $3.83 \times 10^{-12}$  sec<sup>-1</sup>, is the decay constant for Carbon-14. This results in  $N = 1.14 \times 10^{13}$  with window and  $N = 8.49 \times 10^{12}$  without window, where  $N$  is in atoms of Carbon-14.

But, since a  $7 \times 10^{-8}\%$  tag has already been assumed, this leads to  $N = 1.63 \times 10^{22}$  oil molecules, with a detector window and  $N = 1.21 \times 10^{22}$  oil molecules, without a detector window.

By assuming an oil molecular weight of 484 (DC 704) and the area of a one-meter diameter plate, the following sensitivities result:  $M = 1.67 \times 10^{-3}$  g/cm<sup>2</sup> with window and  $M = 1.24 \times 10^{-3}$  g/cm<sup>2</sup> without window.

## B. Plastic Scintillator Analysis

Since this analysis allows the oil to deposit directly on the scintillator, neither a dead layer nor a window is appropriate. The geometry factor would then be one-half and the efficiency would be one, thus,  $A = 100 / (G \cdot E) = 100 / .5 = 200$  dpm.

It is important that the background be minimized when counting at these low levels and techniques of cooling the preamplifier and utilization of coincidence circuitry are considered adequate enough to lower the background to 50 cpm. Thus following the preceding development,  $N = A/\lambda = 8.69 \times 10^{11}$  tagged molecules of oil, which leads to  $M = 1.27 \times 10^{-4}$  g/cm<sup>2</sup> as a detectable limit.

### 2.3 TAGGING OF OILS WITH TRITIUM

The tritium tagging analysis parallels that of Carbon-14. It is again assumed that  $7 \times 10^{-8}\%$  of the oil molecules are tagged with one tritium atom per molecule and the same two detection systems are considered.

#### A. Avalanche Detector Analysis

The development is the same as for Carbon-14. The only difference is the reduced efficiency due to the lower energy beta particles emitted. For the detector with window,  $E = 0.067$  and without window,  $E = 0.617$ .

This results in  $A = 2.83 \times 10^4$  dpm with detector window and  $A = 3.07 \times 10^3$  dpm without window.

Using a  $\lambda$  for tritium of  $1.07 \times 10^{-7} \text{ min}^{-1}$ , the number of molecules of oil,  $N$ , for a window is  $3.78 \times 10^{20}$  and that with no window is  $4.10 \times 10^{19}$ . Converting this to grams of oil and dividing by the area of one-meter diameter plate, gave detectable amounts of  $M = 3.86 \times 10^{-5} \text{ g/cm}^2$ , with a window and  $M = 4.19 \times 10^{-6} \text{ g/cm}^2$ , without a window.

#### B. Plastic Scintillator

Repeating the above analysis, but with  $G$  now equaling one-half and  $E$  equaling unity, an activity of 200 disintegrations per minute results. This yields a value for  $N$  of  $2.68 \times 10^{18}$  oil molecules and a  $M$  of  $2.74 \times 10^{-7} \text{ g/cm}^2$  on a one-meter diameter plate.

### 2.4 SODIUM AND BROMINE TAGGING OF VARIOUS OILS

A previous monthly report indicated trace amounts of Bromine and Sodium in several oils. It was of interest to determine the detectable amounts of oil that could be found

by tagging these oils.

Assume that one irradiates one gram of oil containing one part per million of Sodium or Bromine, which results in  $1 \times 10^{-6}$  grams of Na or Br. Then,  $N_r = N_o \sigma \phi (1 - e^{-\lambda t})$  where  $N_r$  is the number of radioactive atoms produced and  $N_o$  is the number of atoms initially present. For a one microgram mass,  $N_o(\text{Na}) = 2.619 \times 10^{16}$  atoms and  $N_o(\text{Br}) = 7.534 \times 10^{15}$  atoms. The thermal neutron ( $n, \gamma$ ) cross section is represented by  $\sigma$  and the following values were used,  $\sigma(^{23}\text{Na}) = .4167 \times 10^{-24} \text{ cm}^2$ , and  $\sigma(^{79}\text{Br}) = 2.444 \times 10^{-24} \text{ cm}^2$  (Ref. C-12). The neutron flux,  $\phi$ , was assumed to be  $5 \times 10^{12} \text{ n/cm}^2\text{-sec}$  and the values for the decay constant,  $\lambda$ , were  $\lambda(\text{Na}) = 4.621 \times 10^{-2} \text{ hr}^{-1}$  and  $\lambda(\text{Br}) = 1.568 \times 10^{-1} \text{ hr}^{-1}$ . An exposure time, of one hour was chosen as a compromise between maximizing induced activation and minimizing radiation degradation of the oil. Remembering the  $\text{Br}^{79}$  is 50.54% of natural Bromine, the following values of the number of radioactive atoms,  $N_r$ , result:  $N_r(\text{Na}^{24}) = 2.464 \times 10^3 \text{ atoms}/\mu\text{gr Na}$  and  $N_r(\text{Br}^{80}) = 6.753 \times 10^3 \text{ atoms}/\mu\text{gr Br}$ . The activity present then,  $A = \lambda N_r$ , where  $A(\text{Na}^{24}) = 2.719 \text{ dpm}/\mu\text{gr Na}$  and  $A(\text{Br}^{80}) = 17.65 \text{ dpm}/\mu\text{gr Br}$ . Assuming the 100 cpm detected with the previously discussed plastic scintillator to be sufficient for detection, then one obtains as detectable amounts of Na and Br:  $D(\text{Na}) = 1.226 \times 10^{-3} \text{ g Na/cm}^2$  and  $D(\text{Br}) = 1.727 \times 10^{-4} \text{ g Br/cm}^2$ .

Table 1 gives the amounts of Na and Br in parts per million found in several samples of oil.

Table 2-1. PPM of Na and Br in Various Oils

<u>Oil</u>	<u>Na</u>	<u>Br</u>
DC 705	25	0
DC 11	1106	0
Sun Vis 706	43	0
3M-Pt 11	20	39
Hs 1020	50	0
Hs 1055	25	0

Dividing these values into the detectable limits, gave the amounts of oil in grams/cm<sup>2</sup> that could be detected, Table 2.

Table 2-2. Detectable Limits for Tagging of Na and Br

<u>Oil</u>	<u>Na Tag</u>	<u>Br Tag</u>
DC 705	$2.94 \times 10^{-3}$	0
DC 11	$6.65 \times 10^{-5}$	0
Sun Vis 706	$1.71 \times 10^{-3}$	0
3M-PT 11	$3.68 \times 10^{-3}$	$2.91 \times 10^{-4}$
HS 1020	$1.47 \times 10^{-3}$	0
Hs 1055	$2.94 \times 10^{-3}$	0

## 2.5 SILICON TAGGING

Since most of the oils used in vacuum technology are silicon based, silicon tagging was considered as a possible detection method. As the one-meter diameter, cooled, plastic, scintillator detector had offered the best sensitivity in previous analysis, it was the detection used for silicon study. It should be noted that the assumption of a decay time of but one hour is very optimistic and results in the best sensitivity that can be achieved for this method.

It was assumed that the oil (DC 704) was irradiated for one hour at a flux of  $5 \times 10^{12}$  n/cm<sup>2</sup>-sec. and inserted in the vacuum system one hour later. Assuming one kilogram of vacuum oil was irradiated and the parameters of three silicon atoms per molecule of oil, a cross section of  $0.0019 \times 10^{-24}$  cm<sup>2</sup> for Silicon-30 (Ref. C-12), and a Silicon-30 natural abundance of 3.09%, then, the number of atoms of silicon per kg of oil is given by:

$$N_o = \frac{3(1000) (.602 \times 10^{24}) (.0309)}{438} = 1.27 \times 10^{23}$$

Substituting this value into the activation equation and using the parameter values given above and a  $\lambda$  value of  $.265 \text{ hr}^{-1}$  yields:

$$N = N_o \sigma \phi (1 - e^{-\lambda t_1}) e^{-\lambda t_2} = 2.16 \times 10^9 \text{ atoms } {}^{30}\text{Si/kg oil.}$$

This is equivalent to an activity,  $A_1 = N\lambda$ , of  $9.51 \times 10^5 \text{ dpm/kg}$ . As before, 100 cpm was assumed necessary for detection and since  $G = .5$  and  $E = 1$ , the minimum acceptable activity,  $A_2$ , is 200 disintegration/minute.

$$M = A_2/A_1 = 200/9.51 \times 10^5 \text{ dpm/dpm/kg}$$

$$M = 2.67 \times 10^{-5} \text{ gr/cm}^2.$$

## 2.6 FISSION PRODUCT AND ALPHA RANGE--DESCRIPTION

As the results to date had not yielded a detection method with adequate sensitivity, the scope of the study was expanded to include any detection method which used stable or radioactive isotopes. One of the first concepts studied under this enlarged scope was that of using charged particle range to determine total organic gas phase contamination. In order that one may better follow the sensitivity analysis of the subsequent sections, a brief description of the physical layout for this system is given below.

The general design for the range technique consisted of a source with two tubes extending out from it, a variable absorber, and two detector systems placed one each at the ends of the two long tubes, opposite the source. Both detectors are movable and both track evenly. One tube is open to the atmosphere of the chamber, whereas, the other tube, filled with clean air, is sealed and has an exhaust check valve and a bellows for pressure equalization with the chamber. The detectors are placed so that the closed tube is at the proper calibration count point (maximum of  $dI/dX$ ). The difference in count rates allows determination of contamination in the chamber.

## 2.7 FISSION FRAGMENT RANGE

The differential energy loss,  $dE/dX$ , was calculated for fission fragments by (ref. D-42).

$$-(dE/dX) = c/dE^{n/2}. \quad \text{Eq. 1}$$

In the above equation,  $n$  was calculated from  $(\ln(n))/Z^2 = -1.6 - .0533Z$ , where  $Z$  is the average atomic number of the absorbing medium. It is easy to see that for large  $Z$  ( $Z < 10$ )  $n$  approaches zero and  $-(dE/dX) = c/d$ ; where  $x_m$  is the total range,  $E_0$  is the energy of the fission fragment energy,  $d$  is the density, and  $c$  is a constant, fitted by empirical means.



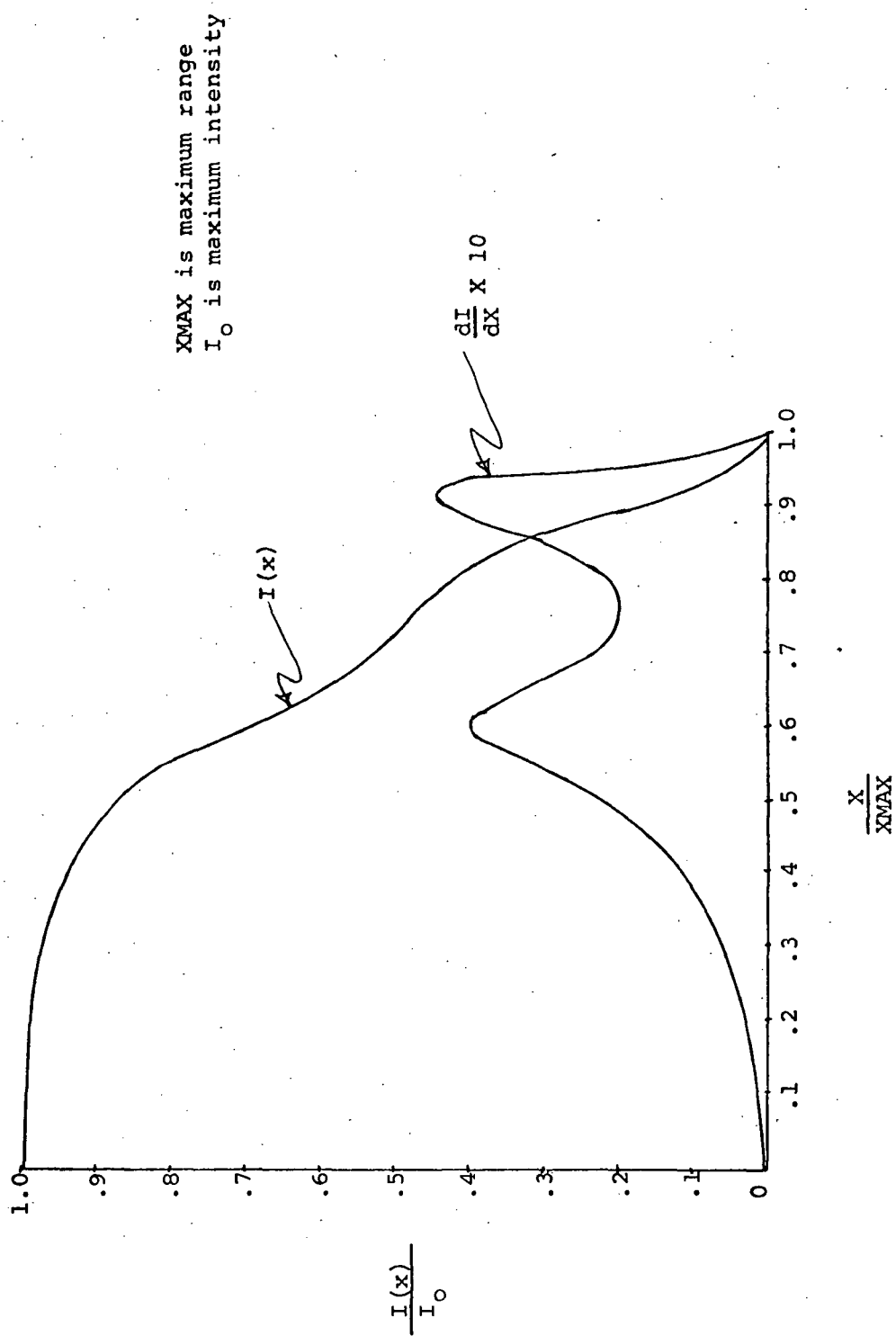
Integrating the energy spectrum of fission fragments (Ref. D-43), beam intensity as a function of energy is obtained. By using equation (1), this energy distribution was converted to a range distribution to get intensity of the beam of fission fragments as a function of range and also  $dI/dX$ .

The curves given, Figure 2-1, are for  $CO_2$ , however, similar properties would be obtained for air. The beam strength entering both tubes was adjusted by addition of absorbers until its magnitude was  $.1 I_0$  and  $dI/dX$  was  $.0438 I_0 / (mg/cm^2)$  where  $I_0$  is the maximum particle intensity and  $dI/dX$  is the rate of decrease in count rate with increasing absorber thickness.

Assuming a pressure of  $10^{-7}$  torr and a temperature of  $20^\circ C$  gives a density for pure air of  $1.85 \times 10^{-7} mg/cm^3$ . To achieve the high count rate necessary for the required statistical accuracy, the calculations assumed a 10 milligram  $^{252}Cf$  source, placed, at the end of the two tubes, with Si(Li) detectors at the other end. Since  $A = N\lambda$ , 10 mg of  $^{252}Cf$  has an activity of  $1.59 \times 10^{12}$  dpm. For the assumed detector system radius of 5 cm., the geometry factor is  $G = \sin^2(0.5 \theta) = 6.25 \times 10^{-6}$ . This gave a fission product beam at the detector of  $1.00 \times 10^6$  particles per minute.

From Figure 2-1 and previous discussion,  $.1 I_0$  is then  $1.00 \times 10^5$  counts for one minute. Counting for 10 minutes gave  $10^6$  counts. For statistical significance, it was required that a difference of at least 1,000 counts exists, or a relative difference of .001. Based on the above, equation 2 represents the intensity relationship developed.

Fig. 2-1. INTENSITY OF FISSION PRODUCT BEAM AS A  
FUNCTION OF DISTANCE TRAVELED ( $Z > 10$ )



$$(.1 I_0 - I_{DC \ 704}) / .1 I_0 = .001 \quad \text{Eq. 2}$$

$$I_{DC \ 704} = .0999 (I_0)$$

Since the reference count rate is  $.1 I_0$ , the difference is  $\Delta I = 1 \times 10^{-4} I_0$ . Using the maximum value of  $\Delta I / \Delta X$ ,  $.0438 I_0$ , the corresponding value of  $\Delta X$  is obtained,  $\Delta X = 2.29 \times 10^{-6} \text{ g/cm}^2$ . This is equivalent to a maximum sensitivity of  $2.29 \times 10^{-9} \text{ g/cm}^3$  for a 10 meter tube length.

As the previous development implies, the sensitivity obtained is linearly dependent on tube length. Thus a 5 meter tube would have a minimum sensitivity of  $4.6 \times 10^{-9} \text{ g/cm}^3$  rather than  $2.3 \times 10^{-9} \text{ g/cm}^3$  as calculated for the 10 meter tube length.

For the situation of molecular flow, it is obvious that the response of the system would also depend on the orientation of the system with respect to the flow path. For purposes of the calculations the condition resulting in maximum sensitivity has been assumed; that the detector was oriented in the center of the molecular flow path and parallel to it.

## 2.8 ALPHA RANGE IN A VACUUM

The development for alpha range follows that which was previously presented for fission fragment range. The source considered in this case was  $^{241}\text{Am}$  and the detector system remained the same.

From Reference C-13, the maximum beam attenuation of an alpha particle beam occurs at 1 MeV and is

$$\frac{dI}{dX} = \frac{-2.03 I_0}{\text{mg/cm}^2} \quad \text{Eq. 3}$$

where,  $I_0$  is in particles per second (Figure 2). The density of air at 20°C and  $10^{-7}$  torr is  $1.85 \times 10^{-7} \text{ mg/cm}^3$ . Assuming a 10 meter tube gave a  $\Delta X$  value of  $1.85 \times 10^{-4} \text{ mg/cm}^2$  and thus,  $\Delta I$  value of  $(-2.03 I_0) (1.85 \times 10^{-4}) = -3.76 \times 10^{-4} I_0$ .

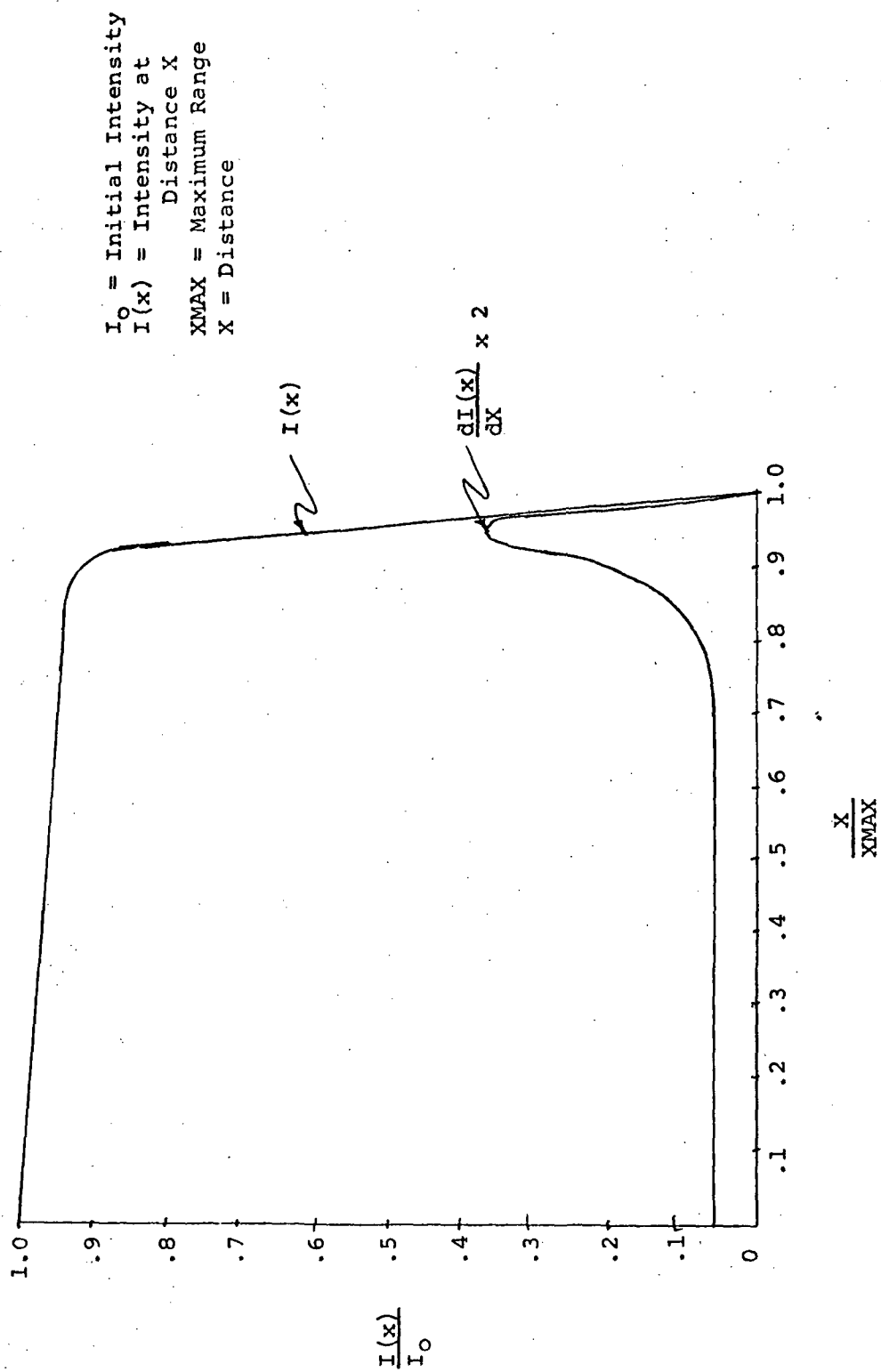
A statistical precision in relative difference of counts of 0.1% was assumed achievable and used in the detection limit equations. As the detection sensitivity is linearly dependent on this percentage, a value of 1% would result in a detection limit a factor of 10 higher than that calculated below. It then followed that the minimum  $\Delta I_2$  that could be found was given by Equation 4.

$$\frac{\Delta I_2 + 3.76 \times 10^{-4} I_0}{-3.76 \times 10^{-4} I_0} = .001 \quad \text{Eq. 4}$$

$$\Delta I_2 = -3.76 \times 10^{-7} I_0$$

It was assumed that oil acts in the same manner as does air in stopping alpha particles and that the oil was dispersed in dry air, then, the change in beam intensity due to the oil was  $\Delta I(\text{oil}) = \Delta I_2 = \Delta I$  or  $\Delta I(\text{oil}) = -3.8 \times 10^{-7} I_0$ . This corresponded to a minimum detectable absorber limit,  $\Delta X$ , of  $1.85 \times 10^{-10} \text{ g/cm}^2$ . It should be noted that this value is the detectable limit whether dispersed or plated upon absorber or detector and, if dispersed, would represent a density of oil in the 10-meter tube of  $1.85 \times 10^{-13} \text{ g/cm}^2$ .

Fig. 2-2. INTENSITY OF ALPHA PARTICLE BEAM AS A  
FUNCTION OF DISTANCE TRAVELLED



## 2.9 NON-RADIOACTIVE ISOTOPE TECHNIQUES

Some consideration was given to using stable isotopes, namely, Carbon-13 and Deuterium, as a means for determining the contamination of a particular oil. Two methods of analysis were considered: Nuclear Magnetic Resonance and Mass Spectrometry. Information concerning these two methods was obtained by consulting members of the Texas A&M University staff currently active in these fields.

### A. Nuclear Magnetic Resonance (NMR) (Ref. C-14).

This technique was deemed infeasible due to the fact that at least 10 mg are necessary for analysis and that the maximum sensitivity was approximately  $10^{-3}$  g of  $C^{13}$  or  $H^2$ , which if accumulated on a  $1\text{ m}^2$  plate, yields only a sensitivity of  $10^{-7}$  g/cm<sup>2</sup>.

### B. Mass Spectrometry

This technique proved impractical due to the large ratio (10% to 20%) of  $^{13}C$  to  $^{12}C$  or  $^2H$  to  $^1H$  required for detection. Again, large, 1 mg samples would be required and the maximum sensitivity of approximately  $10^{-4}$  grams of  $^{13}C$  or  $^2H$  limited the ability of this technique to adequately measure the contamination levels present.

## 2.10 RADIOLOGICAL SAFETY CONSIDERATIONS

Since NASA ground rules (Ref. C-15) for the study precluded the use of isotopes with half-lives greater than 12 hours, Carbon-14 and tritium were now excluded from further consideration. However, the method is quite valid for use

in testing suspect materials in small vacuum systems. The tritium technique would be the preferred method because of its greater sensitivity and lower radiological hazard. The smaller chambers would, however, require a reduction in the surface area of the plastic scintillator from 1 meter in diameter to some more appropriate size and this would reduce the sensitivity somewhat, but any such reduction would be in direct proportion to the ratio of the surface areas between the considered case and the new one.

Of the systems remaining, only the alpha range has the necessary sensitivity to warrant consideration. For the proposed source, Am-241 as a sealed alpha source is acceptable to the NASA Radiological Safety Office and was selected for use. No unacceptable radiological hazard was anticipated, as the decay daughter is Np-237, which has a  $2 \times 10^6$  year half-life, therefore, the decay chain is essentially non-radioactive.

## 2.11 THE ALPHA RANGE APPARATUS

System description: A .73 curie Americium-241 sealed source, activity required to give the necessary count rate, is followed by a set of absorbers. This would then lead to two tubes each 10 meters long and 8 cm in diameter,

placed  $5^\circ$  from the normal of the source absorber box (see Fig. 2-3). One tube would be open and the other sealed with a release valve and a bellows-assembly for pressure normalization with the chamber. The open tube would be sealed at the source end to prevent contaminants from reaching the source box. At the other end of each tube would be a Si(Li) detector mosaic, which could be adjusted in position to obtain maximum sensitivity (see Fig. 2-3). The Si(Li) detector was chosen as it allows the flexibility of counting the number of alphas as well as accurately determining their energy. A possible substitute would be surface barrier solid state detectors. The positioning mechanism would be a set of servos, which would vary source-detector position with the changing pressure. Preamps connected to the Si(Li) detectors would lead pulses to an amplifier-multichannel analyzer assembly outside the vacuum chamber walls (Fig. 2-4). All components within the chamber would be operable at  $\text{LN}_2$  temperatures, in fact, the cooling of the detectors would have a beneficial effect on their operations.

System calibration: After assembly, the detectors would be placed at the source ends of the tubes, and the source box evacuated. Then, absorbers would be added until the resultant alpha particle beam had an energy of 1 MeV as



Fig. 2-3. ALPHA RANGE APPARATUS

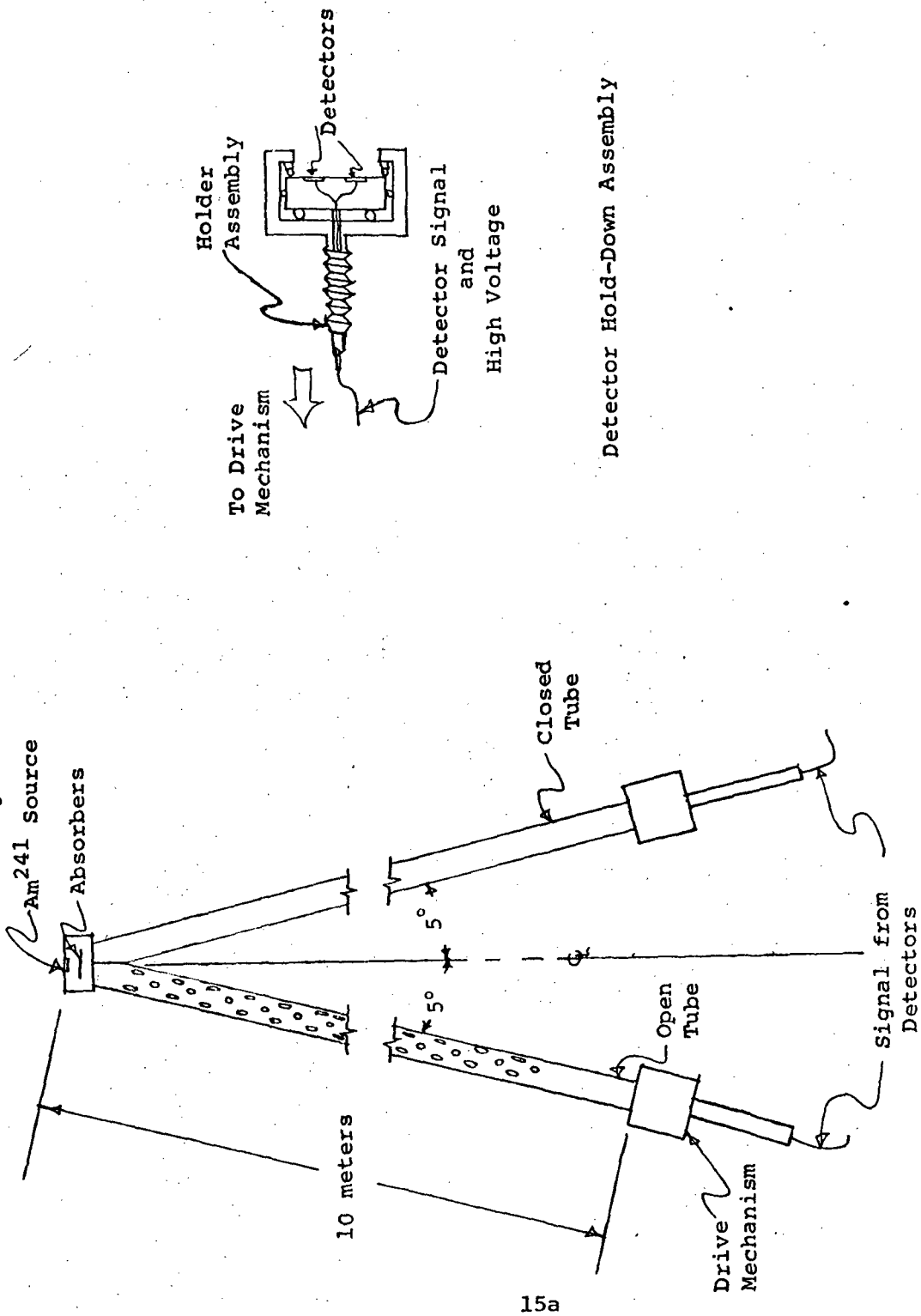
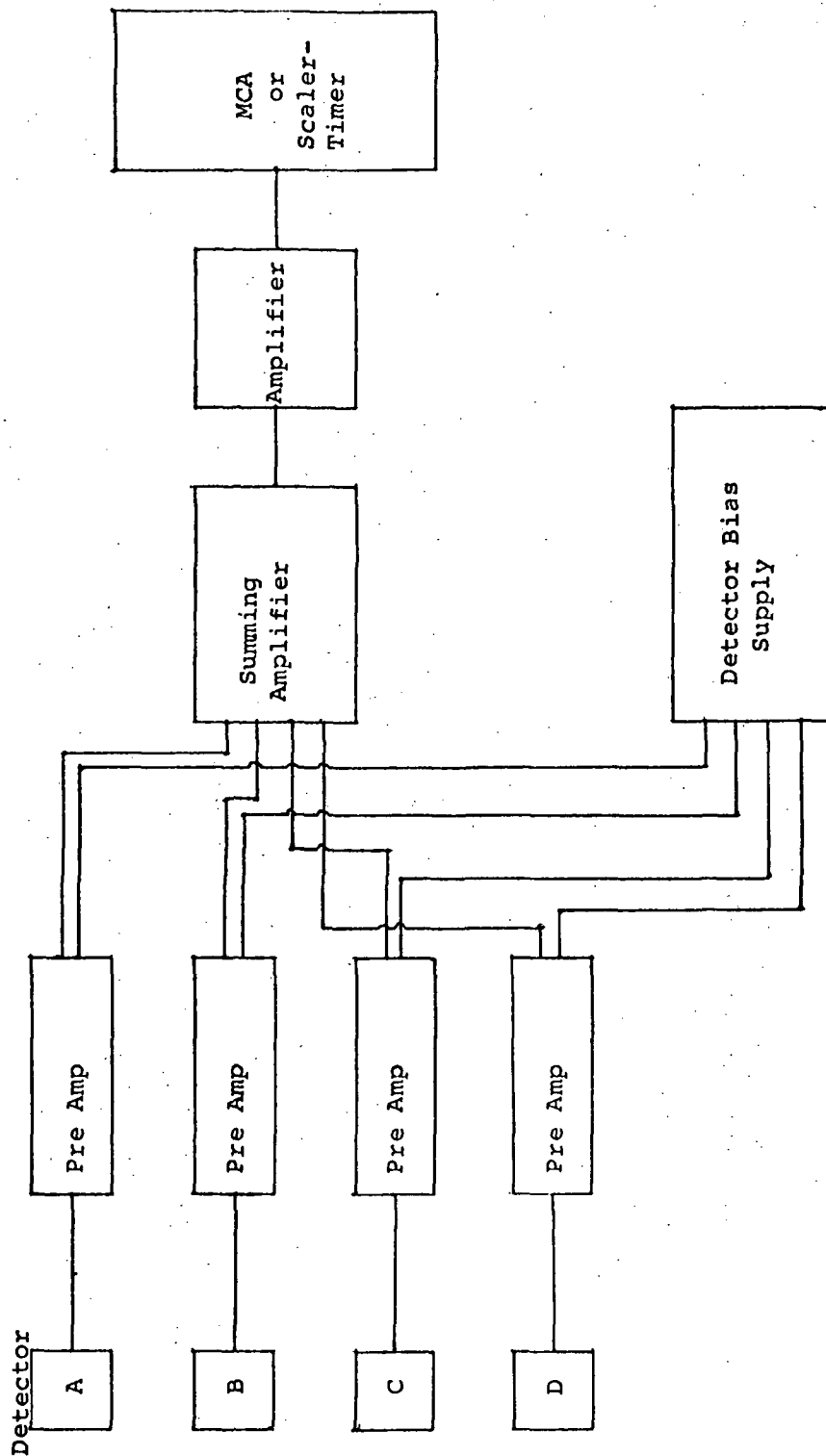


Fig. 2-4. BLOCK DIAGRAM FOR ALPHA RANGE APPARATUS



observed on the multichannel analyzer. Air would then be let into the source box and exact  $dI/dX$  determined. Absorbers are varied until this value becomes a maximum and the open tube amplifier gain would then be adjusted to give the same energy calibration as that of the closed tube system.

System operation: The closed tube and source box are purged with clean, dry air at ambient conditions and the system checked out. As vacuum is achieved, the closed tube detector is placed at the point of maximum  $dI/dX$  and the open tube detector at an equal distance from the  $^{241}\text{Am}$  source. The multichannel analyzer would be placed on the open tube and allowed to take sequential readings in a multiscaler mode and the closed tube detector would be placed on the scaler-timer configuration and run periodically as a control. The contamination level would then be calculated by:

$$\frac{(\text{Count rate open} - \text{count rate closed})}{(dI/dX) (1000)} = g/\text{cm}^3 \quad \text{Eq. 5}$$

The simple relation above, Eq. 5, considers only the oil molecule and neglects the presence of the air molecule in the comparison chamber which equal pressure requires. The error resulting this can be approximated by  $(14/484) 100\%$  or 2.9%.

The pump down and repressurization phases of the chamber cycle would be the most critical period for this system because it would require the servo drive system to rapidly move the detectors over the length of the ten meter tubes to maintain the detectors at the maximum  $dI/dX$  location.

System Components and Cost: Components and associated costs for the alpha range apparatus are given in Table 2-3.

## 2.12 SUMMARY OF ISOTOPE VACUUM MEASUREMENTS

Several systems were considered and are listed in Table 2-4 with their estimated detection limits for comparison with current techniques.

Table 2-3. SYSTEM COMPONENTS AND COST

<u>Component</u>	<u>Manufacturer</u>	<u>Unit Cost</u>	<u>Total Cost</u>
Tubes, vent system, box, Servo	Custom	\$4,000	\$4,000
2 Amplifiers	Ortec 451	450	900
2 Scalers	Tennelec 540	350	700
1 Timer	Tennelec 541	225	225
1 Multichannel Analyzer	Northern Scientific	5,000	5,000
Teletype Output		1,500	1,500
2 Detector Bias Supplies	Ortec 210	810	1,620
8 Detectors	Ortec R-080-2000-100	1,900	15,800
8 Preamplifiers	Ortec 121	225	1,800
2 Nim Bin	Ortec 401A/402B	540	1,080
2 Sum-Invert Amplifiers	Ortec 433	185	370
Miscellaneous Equipment			
Am-241 Source		1,000	1,000
			<u>3,000</u>
	Total		\$36,995

Table 2-4. SUMMARY OF ISOTOPE VACUUM MEASUREMENTS

Method	Detection Limit	Advantages	Deficiencies
1) CCU <sup>a</sup>	$1 \times 10^{-9}$ g/cm <sup>2</sup>	Simple, versatile, inexpensive; state of the art; in use at present.	Subject to repressurization problems; after the fact type of test.
2) Quartz Microbalance	$1 \times 10^{-9}$ g/cm <sup>3</sup>	Operational over complete chamber cycle; state of the art; presently in use.	Measures only condensable impurities; difficult to achieve published sensitivity of $1 \times 10^{-9}$ g/cm <sup>3</sup> .
3) <sup>14</sup> C tag	$1.7 \times 10^{-3}$ (b) g/cm <sup>2</sup> $1.3 \times 10^{-4}$ (c) g/cm <sup>2</sup>	Operational over complete system; allows selective testing of suspected agents on an individual basis.	Long half-life radioactive contamination of chamber; tag may not follow more volatile fraction of compound.
4) <sup>3</sup> H tag	$4.2 \times 10^{-6}$ (b) g/cm <sup>2</sup> $2.7 \times 10^{-7}$ (c) g/cm <sup>2</sup>	Operational over complete system; allows selective testing of suspected agents on an individual basis.	Long half-life radioactive contamination of chamber; tag may not follow more volatile fraction of compound.

Method	Detection Limit	Advantages	Deficiencies
5) Na tag	$1.71 \times 10^{-3}$ (c) g/cm <sup>2</sup>	Selective tagging of some specific agents; operational over complete cycle.	Tag may not follow more volatile fraction of compound.
6) Br tag	$2.91 \times 10^{-4}$ (c) g/cm <sup>2</sup>	Selective tagging of some specific agents; operational over complete cycle.	Tag may not follow more volatile fraction of compound
7) Si tag	$2.67 \times 10^{-3}$ (c) g/cm <sup>2</sup>	Selective tagging of some specific agents; operational over complete cycle; also short half-life isotope.	Severe time constraint on use of method.
8) Alpha range	$1.85 \times 10^{-10}$ (d) g/cm <sup>2</sup> $1.85 \times 10^{-13}$ (e) g/cm <sup>3</sup>	Fully operational over complete cycle; detects both condensable and non-condensable contaminants; system can be made very directional.	Size problems; sensitivity very directional dependent; new system never before used for this purpose.

Method	Detection Limit	Advantages	Deficiencies
9) F.P. Range	2.29 x 10 <sup>-6</sup> (d) g/cm <sup>2</sup>	Fully operational over complete cycle; detects both condensable and non-condensable contaminants; system can be made very directional.	Size problems; sensitivity very directional dependent; new system never before used for this purpose. Also, possible radioactive contamination problem

- a -- Method currently used by NASA
- b -- Avalanche detector system
- c -- Plastic scintillator system
- d -- Oil plated on source or detector
- e -- Oil dispersed in dry air at 10<sup>-7</sup> torr and 20°K



### 3.0 ACTIVATION ANALYSIS (PHASE II)

The research objective of this phase of the project is to evaluate the state-of-the-art and determine the feasibility of using nuclear activation analysis to identify and locate sources of contamination in the NASA space environmental simulation chambers. Several activation analysis alternatives were investigated on the basis of technical feasibility, expected performance and cost. Efforts were directed primarily at neutron activation analysis techniques for detection of several oils, greases, and paint which were suspected as likely contaminants. Charged particle and high-energy photon activation analysis techniques were not included in the studies due to the high cost of particle accelerators and the lack of detailed experimental data on which to base sensitivity estimates.

The basic approach to the problem of contaminant identification with neutron activation analysis involves detecting a specific element or set of elements whose presence in a sample uniquely identifies a given contaminant and whose concentration in a sample is directly proportional to the amount of that contaminant present. Since activation analysis is an elemental method incapable of determining compounds or molecular structure, this approach requires

that the elemental composition of each possible contaminant contain a unique set of elements suitable for neutron activation analysis. This unique "fingerprint" characteristic may be an inherent property of the material or may be the result of a process in which certain suitable elements are placed into the compound for use as "tag" elements.

During this investigation, emphasis was placed on the following six materials commonly used in the space environmental simulation chamber and which were thought to be likely contaminants:

Dow Corning	705
Dow Corning	11
Sun Vis	706
3-M Black Velvet Paint	
Houghton Safe	1020
Houghton Safe	1055

### 3.1 BASIC DESCRIPTION OF METHOD

Neutron activation analysis can be generally described as having three basic steps:

1. sample activation
2. sample cooling
3. gamma ray spectrometry

Sample activation consists of irradiating a material with a flux of neutrons to produce radioisotopes by means of nuclear transmutation. Sample cooling is an optional step which may be used to allow undesirable short lived radio-

isotopes to decay to an acceptable activity level before attempting to measure the activities of the elements of interest. After suitable cooling period, gamma ray spectrometry is used to measure the energies and intensities of the various gamma rays emitted by the activated material. Using the spectral data, a comparison can be made between the spectra from an unknown sample and those from a known standard to provide identification of elemental constituents and a determination of the elemental concentrations. Under optimum conditions with high neutron flux densities, over half of the naturally occurring elements can be detected in quantities below one microgram (I-14).

### 3.2 ACTIVATION ANALYSIS OF POTENTIAL CONTAMINANTS

Samples of the six potential contaminants were evaluated on the basis of their elemental composition to determine their suitability for detection and identification using neutron activation. The major elements comprising these materials include carbon, hydrogen, oxygen, silicon and phosphorus, none of which activate appreciably with thermal neutrons and only silicon, oxygen and phosphorus activate well with fast neutrons. Fast neutron activation analysis was considered as a possible technique but rejected due to

lack of a practical means of producing very high fluxes of fast neutrons required for sub-microgram measurement sensitivities. Since their major elements could not be used for "fingerprinting" the potential contaminants, analyses were carried out on samples of these materials to determine if minor or trace elements suitable for thermal neutron activation were present. Analyses were performed at Texas A&M with the Nuclear Science Center reactor which produces a usable thermal neutron flux of  $5 \times 10^{12}$  n/cm<sup>2</sup>/sec. Gamma ray spectrometry was accomplished with a Ge(Li) detector and a 4096 channel pulse height analyzer. Results of these analyses, shown on Table 3-1, revealed no "fingerprint" elements in these samples which would be sufficiently reliable and unambiguous for use in contaminant identification. While several samples showed possible unique trace element patterns, such as hafnium in Houghton Safe 1020 and tungsten in Sun Vis 706, the low concentrations (sub-ppm) make detection of microgram amounts of samples virtually impossible by neutron activation analysis.

Table 3-1 Elemental Analyses of Suspected Contaminants<sup>1</sup>

Sample Element	Concentrations in ppm					
	SUN VIS 706	DC 705	DC 11	3m PT.II	HS 1020	HS 1055
Al <sup>2</sup>	25.0	537.0	1400.00	---	108.0	123.0
Br	0.49	0.52	---	39.0	5.0	1.8
Cl	28.0	15.0	35.0	52,000.0	118.0	313.0
Cu	2.8	0.32	13.0	0.64	1.2	0.47
I	1.9	---	---	---	---	---
La	---	0.08	---	---	---	0.06
Mn	---	---	0.90	---	---	---
Na	43.0	25.0	1106.0	20.0	50.0	25.0
V	0.19	---	0.63	---	---	---
W	0.11	---	---	---	---	---
Zn	168	---	2.4	2.7	1.3	---
Co	---	---	0.2	0.16	---	0.09
Hf	---	---	---	---	0.39	---

<sup>1</sup>Analyses performed using instrumental neutron activation

<sup>2</sup>Al data not corrected for Si interference due to <sup>28</sup>Si(n,p)<sup>28</sup>Al.

### 3.3 INACTIVE TRACERS

When a material is inherently free of suitable "fingerprint" elements, it is often possible to label or tag the material with an element or set of elements which can be measured with good sensitivity and specificity. This method of inactive or stable tracers offers many of the advantages of radioactive tracers while avoiding the radiological safety and radioactive contamination problems. Tagged materials would be used in the chamber in place of usual materials. Samples of contaminants would then be collected in the test chamber and analyzed by neutron activation to determine the identity and amount of each tag element present.

The sensitivity of this procedure for detecting small quantities of a tag element depends upon the element's activation cross section as well as the energy and intensity of the irradiating neutron flux. Tables 3-2 and 3-3 show the measurement sensitivities for those elements detectable by thermal neutron activation and 14 MeV neutron activation respectively. It should be noted that for reactor thermal neutron activation analysis under the conditions defined on Table 3-2, the median detection limit is approximately  $10^{-8}$  grams whereas 14 MeV NAA provides a median detection

Table 3-2 Summary of Experimental Detection Limits  
Determined with Reactor Thermal Neutrons<sup>1</sup>

Atomic No. Range	Minimum detection ranges (grams)										
	10 <sup>-11</sup>	10 <sup>-10</sup>	10 <sup>-9</sup>	10 <sup>-8</sup>	10 <sup>-7</sup>	10 <sup>-6</sup>	10 <sup>-5</sup>	10 <sup>-4</sup>	10 <sup>-3</sup>	10 <sup>-2</sup>	
8 to 9					F					O	
11 to 18		Ar	Na, Al	Cl	Mg	Si, S					
19 to 35	Mn	Sc, V Br	Co, Cu Ga, As, Se	K, Ti, Zn, Ge	Cr, Ni, Ca	Fe					
37 to 53	In		Sr, Rh, Ag, Cd, Sn, Sb, I	Rb, Zr, Mo, Ru, Pd, Te	Y, Nb						
55 to 56		Cs, Hf	Ba, W	Ta, Pt, Os	Pb						
57 to 71	Eu, Dy	Sm, Ho Er	La, Nd Gd, Yb, Lu	Ce, Pr, Tb	Tm						
72 to 82	Au	Re, Ir	Hg								

1. Experimental Conditions:

Irradiation Time = 1.0 hour

Flux =  $4.3 \times 10^{12} \text{ n} \cdot \text{cm}^{-2} \cdot \text{sec}^{-1}$

Detector: 3" x 3" NaI(Tl)

Counting Time: up to 1 hour

Table 3-3. Summary of Measurement Sensitivities Using 14 MeV Neutron Activation<sup>1</sup>

Atomic No.	Sensitivity Range (grams) <sup>2</sup>				
	10 <sup>-5</sup>	10 <sup>-4</sup>	10 <sup>-3</sup>	10 <sup>-2</sup>	10 <sup>-1</sup>
3 - 10		Al, F, B, N, O			
11 - 18	P, Si	Na	Cl, Mg		
19 - 36	Br, Cr, Cu	Cd, Rb, Ga, Se, Ge, Mn, Ni	As, Co, Fe, K, Ti		
37 - 54	Ag	Sb, Pd, Sr, Sn, In	Mo, Ru,	Nb	
55 - 86	Ba, Ce, Pr,	Dy, Au, Hg, Ta, Tb	Cs, Nd, Pb, Sm, Lu, Er, Eu, Hf	Ir, Gd	

<sup>1</sup>Experimental Conditions:

Irradiation time = min {5 min, 3t<sub>1/2</sub>}

Flux = 2.0 x 10<sup>8</sup> n · cm<sup>-2</sup> · sec<sup>-1</sup>

Delay Time = min {1 min, 1t<sub>1/2</sub>}

Counting Time = min {5 min, 3t<sub>1/2</sub>}

Detector: 2 matched 3" x 3" NaI(Tl) crystals separated by 2.4 cm

<sup>2</sup>After Cuypers (H-8)



limit in the range of  $10^{-3}$  to  $10^{-4}$  grams using a state-of-the-art Cockcroft-Walton type neutron generator. It is obvious that for thermal neutron activation, elements with the best sensitivity (Eu, Dy, Mn, etc.) would be desirable for use as tags, however, a further requirement is that the tag element be chemically suitable for inclusion in the compound being labeled.

In view of this chemical compatability requirement, a brief study was undertaken to determine the practicality and reliability of an elemental tagging system. In reference to the chemical makeup of the expected contaminant materials, a typical electrophilic aromatic substitution process (halogenation) was chosen for the tagging mechanics. The elements best suited for this tagging procedure and trace activation analysis are chlorine and bromine. The halogenation is expected to take place on the phenyl radical common to the proposed DC-11, DC-705, HS-1055, and HS-1120 contamination materials. The general equation for this reaction is:  $ARH + X_2 \xrightarrow{\text{Lewis Acid}} ARX + HX$

where:

AR = phenyl radical

$X_2$  =  $Cl_2$  or  $Br_2$

Lewis Acid =  $FeCl_3$ ,  $AlCl_3$ , etc.

### 3.4 MEASUREMENT SENSITIVITY

A primary measure of the effectiveness of a neutron activation analysis system for contaminant detection is the sensitivity with which tag elements can be determined under given conditions of sample acquisition, irradiation, and counting. A large number of variables must be considered in estimating measurement sensitivity, some of which are set by system design and others determined by the physical properties of the materials in the sample. The more important factors which influence system measurement sensitivity are given on Table 3-4.

In order to assess the feasibility of using bromine, chlorine and iodine as tag elements to be detected by a neutron activation analysis system located in or near the chamber, a set of "best case" conditions were assumed and detection limits calculated. First, detection limits for elemental Br, Cl and I were calculated as described in Appendix B and results shown in Table 3-5. These detection limits are based upon a one hour irradiation with a thermal neutron flux of  $1.25 \times 10^9$  n/cm<sup>2</sup>/sec and counted with a 3 in. x 3 in. NaI(Tl) detector. Further assumptions which were made to determine "best case" performance are as follows:

Table 3-4. PARAMETERS AFFECTING MEASUREMENT SENSITIVITY  
WITH NEUTRON ACTIVATION ANALYSIS

I. SAMPLE CONDITIONS

- (1) Isotopic abundance of chosen isotope.
- (2) Maximum size of sample suitable for irradiation.
- (3) Nuclear activation properties of the other constituents in sample.

II. IRRADIATION CONDITIONS

- (1) Activation cross section for chosen reaction.
- (2) Available flux.
- (3) Irradiation time.
- (4) Half life of activation product.

III. POST-IRRADIATION PROCESSING

- (1) Decay during elapsed time from end of irradiation to start of counting.

IV. RADIATION MEASUREMENT

- (1) Detector background.
- (2) Minimum detectable counting rate above background.
- (3) Energy resolution of the system, FWHM.
- (4) Over-all counting efficiency for the radionuclide.

Table 3-5. TAG ELEMENTS

TAG ELEMENT	HALF LIFE	$\gamma$ ENERGY (MeV)	CROSS SECTION (barns) <sup>a</sup>	DETECTION LIMIT <sup>d</sup> ( $\mu$ gms)	NOMINAL CONTAMINANT TAG LEVEL <sup>b</sup> ( $\mu$ gms)	R FACTOR <sup>c</sup>
BROMINE <sup>80</sup> Br	4.4hr	.62	6.7	2.33	17.29	7.4
CLORINE <sup>38</sup> Cl	37.3m	1.64	33.8	9.3	10.01	1.1
IODINE <sup>128</sup> I	25m	.455	7.0	2.74	22.75	10.0

(a) Thermal Absorption Cross Section

(b) Deposited on 45.58 cm<sup>2</sup> collection disk

(c) Reliability Factor

(d) 1 hour irradiation in thermal neutron flux of  $1.25 \times 10^9$  n/cm<sup>2</sup>-sec

1. Tag element is uniformly distributed in the contaminant material.
2. Only one contamination source is responsible for the tagged material deposited on the 3 inch diameter collection disk.
3. Tag element does not disassociate from the contaminant materials.
4. Average contaminant deposition is approximately  $10^{-6}$  gm/cm<sup>2</sup>.
5. A non-activable collection medium is used in lieu of presently used pyrex plate.

Assumptions one through three above are "best case" conditions with respect to the measurement of trace element tags in chamber contaminant samples. To assure that the first three assumptions are valid would require further research beyond the scope of this project; however, it should be pointed out that variations in any one of these would significantly degrade the overall effectiveness of the tagging approach. Assumption number three (stability of tag element in the contaminant material) is probably the most uncertain of all. The effects of extreme temperatures and high vacuum could cause the release of tag elements from the labelled material thus defeating the method. A quantitative evaluation of this factor would require extensive testing of various tagged contaminants under conditions equivalent to those present in the test chamber. Number four assumes a contaminant

deposition of  $1 \times 10^{-6}$  g/cm<sup>2</sup> which approximates levels observed in previous tests at sampling sites behind the chamber heat sink panels and directly in front of diffusion pump ports (J-1).

Assumption number five suggests the replacement of the pyrex CCU with a non-activable collection medium. In evaluating pyrex glass (Corning 7740), it is clear that the sodium and potassium content (3.8% and 0.4% by weight, respectively) would activate with neutrons to produce radioactivity levels sufficiently high to mask the expected activity from the tag elements. The use of an analytical grade paper filter would be a sufficient substitute for the pyrex plate if adequate thermal protection is provided.

The last column in Table 3-5 is intended to show the extent to which the proposed technique would exceed minimum required performance under "best case" conditions. The reliability factor, R, is the ratio of the maximum expected amount of tag element over the minimum detectable amount of the tag element and is calculated as follows:

$$\frac{(\% \text{Tag}) \quad (\text{Den}) \quad (\text{Area})}{(\text{Det})}$$

where: %Tag = weight % tag element in contaminants

Den = average contaminant density on collection  
plate ( $\sim 10^{-6}$  gm/cm<sup>2</sup>)

Area = area of sample collection plate (45.5 cm<sup>2</sup>)

Det = detection limit of tag element in micrograms (reference Appendix B)

Examining the "best case" reliability factors and the validity of the assumptions made, it becomes apparent that without a substantial increase in neutron flux over the assumed value of  $1.25 \times 10^9$  n/cm<sup>2</sup>/sec, system performance would be very marginal.

### 3.5 FOUR IRRADIATION ALTERNATIVES FOR NEUTRON ACTIVATION ANALYSIS

The previous section discussed measurement sensitivities and provided system performance estimates based on a neutron flux of  $1.25 \times 10^9$  n/cm<sup>2</sup>/sec. This section describes four alternatives for a NAA system and summarizes their important characteristics in Table 3-6.

The on-line monitoring system, which is the major concern of the next section of this report, is composed of a shielded isotopic neutron source, sample changer, and a NaI(Tl) gamma ray spectrometer located inside NASA Chamber A. The major advantage of this type system is its ability to offer hourly real-time analysis during chamber operation. A major consideration in this approach would be the effect of hostile environmental conditions (ie. 125°C and  $10^{-6}$  Torr vacuum) inside the chamber during operation. Precautions must be taken to seal the shield assembly to prevent outgassing from the hydrogenous source shield.

The second alternative, a variation of the previous approach, involves the use of a subcritical multiplier system which employs a centrally located neutron source in a <sup>233</sup>U-H<sub>2</sub>O or <sup>235</sup>U-H<sub>2</sub>O solution. Such a system, as suggested by Currie, McCrosson and Parks (G-17), could



Table 3-6. SUMMARY OF IRRADIATION SYSTEM CHARACTERISTICS

TECHNIQUE	INITIAL COST ANALYSIS	ANALYSIS SPEED	OBTAINABLE THERMAL NEUTRON FLUX <sup>1</sup> (n/cm <sup>2</sup> -sec)	MEASUREMENT SENSITIVITY
ON-LINE <sup>252</sup> Cf SYSTEM (IN-CHAMBER)	\$525,000 <sup>3</sup>	HOURLY	1.25 x 10 <sup>9</sup>	POOR
<sup>252</sup> Cf AND MULTIPLIER SYSTEM <sup>2</sup> (ON-SITE)	\$200,000 <sup>3,4</sup>	DAILY	1.52 x 10 <sup>9</sup>	FAIR
14 MeV GENERATOR (ON-SITE)	\$25,000	DAILY	4.0 x 10 <sup>7</sup>	VERY POOR
REACTOR AT TEXAS A&M UNIVERSITY	\$600 min. <sup>5</sup>	SEVERAL DAYS	4.3 x 10 <sup>12</sup>	GOOD

- (1) Uniform flux over 3 inch diameter disk.
- (2) 5-3 mg sources with a 4 times multiplier.
- (3) Based on 10 dollars per microgram.
- (4) Cost of multiplier approximately \$50,000.
- (5) \$300/day for reactor time plus \$30/sample for analysis. Minimum 10 samples.

enhance the neutron flux by factors of 6.4 and 4.8 respectively. However, to achieve these useful flux enhancement factors the assembly must be operated so near criticality that safety procedures and regulations may preclude practical application of this approach. Due to the complexity and size of this system, it would most likely be located outside the chamber, offering only daily analysis. This technique is mentioned only for comparison purposes here but will be discussed in more detail in the next section of this report.

The third alternative uses a 14 MeV neutron generator as the source of neutrons. A system of this type would provide a peak thermal neutron flux of approximately  $4 \times 10^7$  n/cm<sup>2</sup>/sec, which is insufficient to provide sensitivities required. This technique is mentioned, as in the previous case, only for comparison purpose.

The last technique proposed involves using the nuclear reactor at Texas A&M University. Although the return time analysis is several days, the available thermal neutron flux of  $4.3 \times 10^{12}$  n/cm<sup>2</sup>/sec offers the best sensitivity and reliability for contaminant identification.

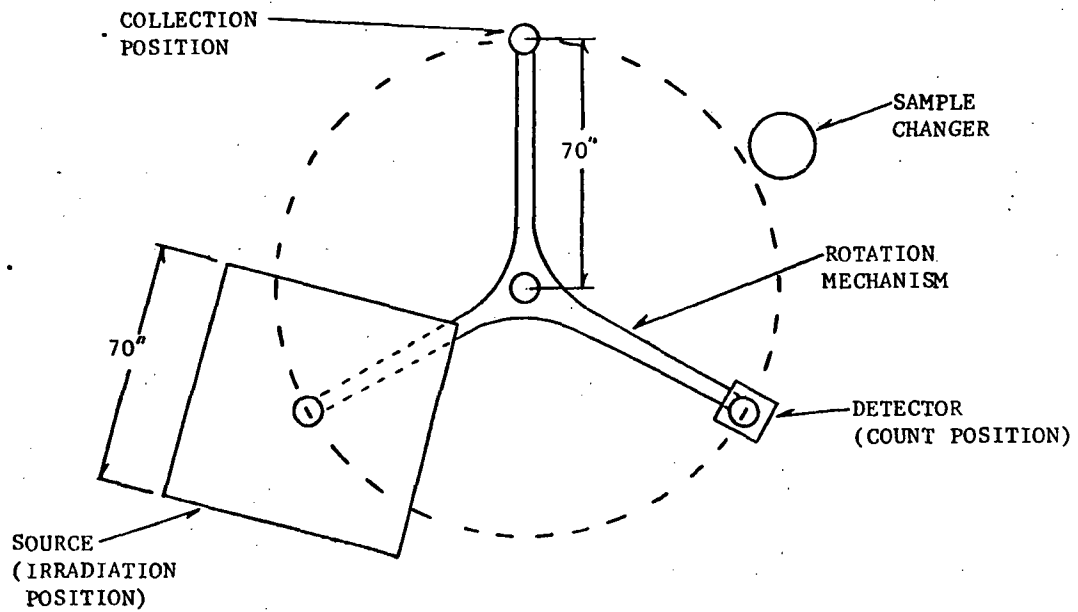
In evaluating the above approaches, one should

compare the costs, analysis speed, and measurement sensitivity to determine the applicability to a given problem.

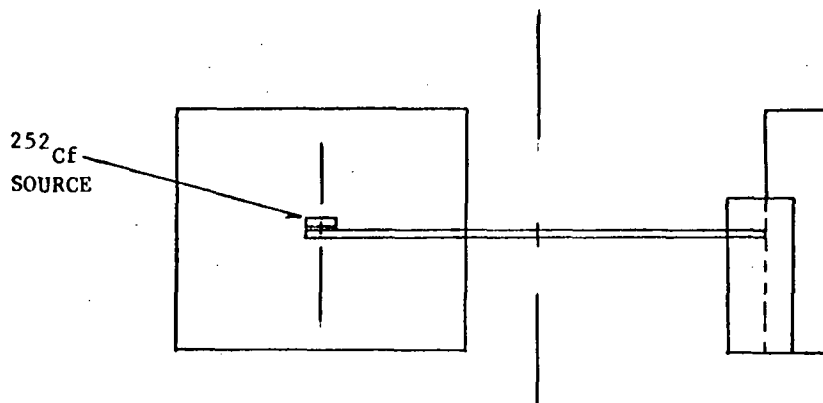
### 3.6 SYSTEM CONFIGURATION

The development of a neutron activation system for contaminant identification depends first on tagging the contaminant or contaminants with suitable elemental tags, and second, on a reliable means of detecting and identifying these tags in collected contamination samples. This section describes a system for accomplishing in-chamber sample collection and analysis by means of an automatic sampling arrangement, an isotopic neutron source, and a scintillation detector. The operation of this system, shown schematically in Figure 3-1, involves a three-arm rotating sample handler which positions sample collection disks in proper position for acquisition, irradiation, counting or changing on a programmed basis. The first position of rest is the collection position where the collection disk is exposed to the chamber atmosphere for an hour. Then a 120 degree counterclockwise rotation places this collection disk under a uniform neutron flux of  $1.25 \times 10^9 \text{ n/cm}^2/\text{sec}$  for an hour irradiation.

Fig. 3-1.  $^{252}\text{Cf}$  ON LINE SYSTEM



TOP VIEW



FRONT VIEW

Upon completion of this phase, the collection disk is rotated another 120 degrees to the NaI detector position where a one-hour gamma spectral analysis is made to determine the type and amount of tag elements present on the collection disk. From this position, the collection disk is rotated 60 degrees where it is replaced with a clean collection disk by the sample changer. Another 60 degree rotation places the new disk in the original position and the process is repeated in 60 degree steps. Having a collection disk at each position provides an hourly analysis of chamber contamination after the initial 2 hour start-up time. Each of these positions will be discussed in greater detail below.

Of prime importance in the design of an in-site neutron activation system is the selection of a suitable neutron source. The neutron source represents the limiting factor relative to overall measurement sensitivity and is the most expensive single item in the system by a vast margin; therefore, its selection must be done carefully. After a comparative analysis of several isotopic neutron sources, Californium-252 was chosen. The distinguishable characteristics of Cf-252

can be summarized in the following two properties:

1. Cf-252 emits a large number of neutrons by spontaneous fission ( $2.34 \times 10^{12}$  n/sec/gm)
2. Cf-252 half-life is reasonably long (2.65 years).

The main advantage that a Cf-252 source has over conventional neutron sources, such as  $^{210}\text{Po-Be}$ ,  $^{241}\text{Am-Be}$  or  $^{238}\text{Pu-Be}$  is its significantly smaller dimensions. Not only is the active source volume smaller in Cf-252, but it also requires less space to accomodate decay helium and does not require the extensive heat-dissipating surfaces required by some ( $\alpha, n$ ) sources. Therefore, the portability of this source is only limited by its shielding requirements. A summary of the neutron source analysis is presented in Table 3-7.

The cost of neutron sources with a total neutron output rate of  $5 \times 10^{10}$  n/sec (needed in activation analysis) are compared in Table 3-8. At an estimated cost of \$1,000,000 per gram for Cf-252, this source offers the most favorable combination of initial investment plus yearly replenishment costs of any isotopic neutron source. As larger quantities of Cf-252 are produced to meet the demand, the price is expected to drop according to the following production level and

Table 3-7. ISOTOPIC NEUTRON SOURCE CHARACTERISTICS

Source <sup>a</sup>	Half-Life	Type	Yield (n/sec/Ci) x 10 <sup>6</sup>	Thermal <sup>c</sup> Reduction Factor	Gamma Dose Rate (r/hr @ 1 m)	Average n Energy (MeV)	Heat Generation (Watts)	Volume <sup>b</sup> (cm)
<sup>124</sup> Sb-Be	60d	γ,n	1.6	10	4.5 x 10 <sup>4</sup>	.024	20	200
<sup>210</sup> Po-Be	138d	α,n	2.5	400	2.0	4.3	640	200
<sup>242</sup> Cm-Be	163d	α,n	4.0	400	0.3	4.0	600	2
<sup>252</sup> Cf	2.65y	Spontan- eous Fission	4400	100	2.9	2.3	0.8	<1
<sup>244</sup> Cm-Be	18.1y	α,n	3.0	400	0.2	4.0	600	70
<sup>238</sup> Pu-Be	89y	α,n	2.8	400	0.4	4.0	550	350
<sup>241</sup> Am-Be	458y	α,n	2.0	400	2.5	4.0	750	2.2 x 10 <sup>4</sup>

(a) Source normalized to 5 x 10<sup>10</sup> n/sec

(b) Not including void space for Helium from α decay

(c) Based on (G-17) in water, pp. IV-28

Table 3-8. ISOTOPIC NEUTRON SOURCE COST ANALYSIS<sup>a</sup>

Source	Cost per $5 \times 10^{10}$ n/sec (Dollars)
$^{242}\text{Cm-Be}$	10,000
$^{210}\text{Po-Be}$	20,000
$^{252}\text{Cf}$	20,000
$^{124}\text{Sb-Be}$	25,000
$^{244}\text{Cm-Be}$	280,000
$^{238}\text{Pu-Be}$	310,000
$^{241}\text{Am-Be}$	1,500,000

(a) Based on (G-1)



price trends:

	<u>Mid 70's</u>	<u>Late 70's</u>	<u>Early 80's</u>
Avg. Quantity (gm/yr)	5-10	20-50	40-250
Price (\$/μ gm)	5-7	2-3	0.5-1.5

It is clear that small microgram quantities of Cf-252 will not provide adequate neutron activation for low yield samples. In order to achieve a neutron flux of  $1.25 \times 10^9$  n/cm<sup>2</sup>/sec, a 50 milligram Cf-252 source is required. In view of the relatively short lifetime (~ 10 years) of Cf-252, upkeep of sources of this magnitude will be extremely costly even with the projected decrease in Cf-252 prices over the next 10 years. One method of increasing the available neutron flux for a given source strength is to use a subcritical neutron multiplier. With this method, multiplications ranging from 10 to 200 are possible with safe, simple and relatively inexpensive devices. Therefore, by spending half the source budget for Cf-252 and half for a multiplier system, the annual upkeep expenditure is reduced by  $\frac{1}{2}$  while the available flux is increased by a factor of 5 to 100 than if the entire budget was used for the purchase of Cf-252. The major drawback of this

system is that it would require an impractical size for on-line analysis in the chamber. It should also be pointed out that more work on optimizing the output and on demonstrating the inherent safety is required before inexpensive licensing of this method is a reality. Miller and Kunze (G-16) have designed an optimum neutron multiplier system shown in Figure 3-2 which should be considered as an alternative in the design of an activation analysis system using Cf-252.

The next area of interest was the design of a source matrix which would optimize the activation of a 3 inch diameter collection disk. Several source arrangements were investigated to determine an adequate activation method. The 5 individual 10 milligram sources in a cluster design, Figure 3-3, provides a high average flux density of  $1.08 \times 10^9$  n/sec/cm<sup>2</sup>. This arrangement was obtained by observing the thermal neutron flux distribution of Cf-252 in water, which shows that the peak thermal flux is about 0.5 inches from the surface of the source (G-18). Although the flux distribution is not a constant over the entire contaminant collection disk, the slightly centrally peaked flux distribution as shown in Figure 3-3 will provide efficient sample

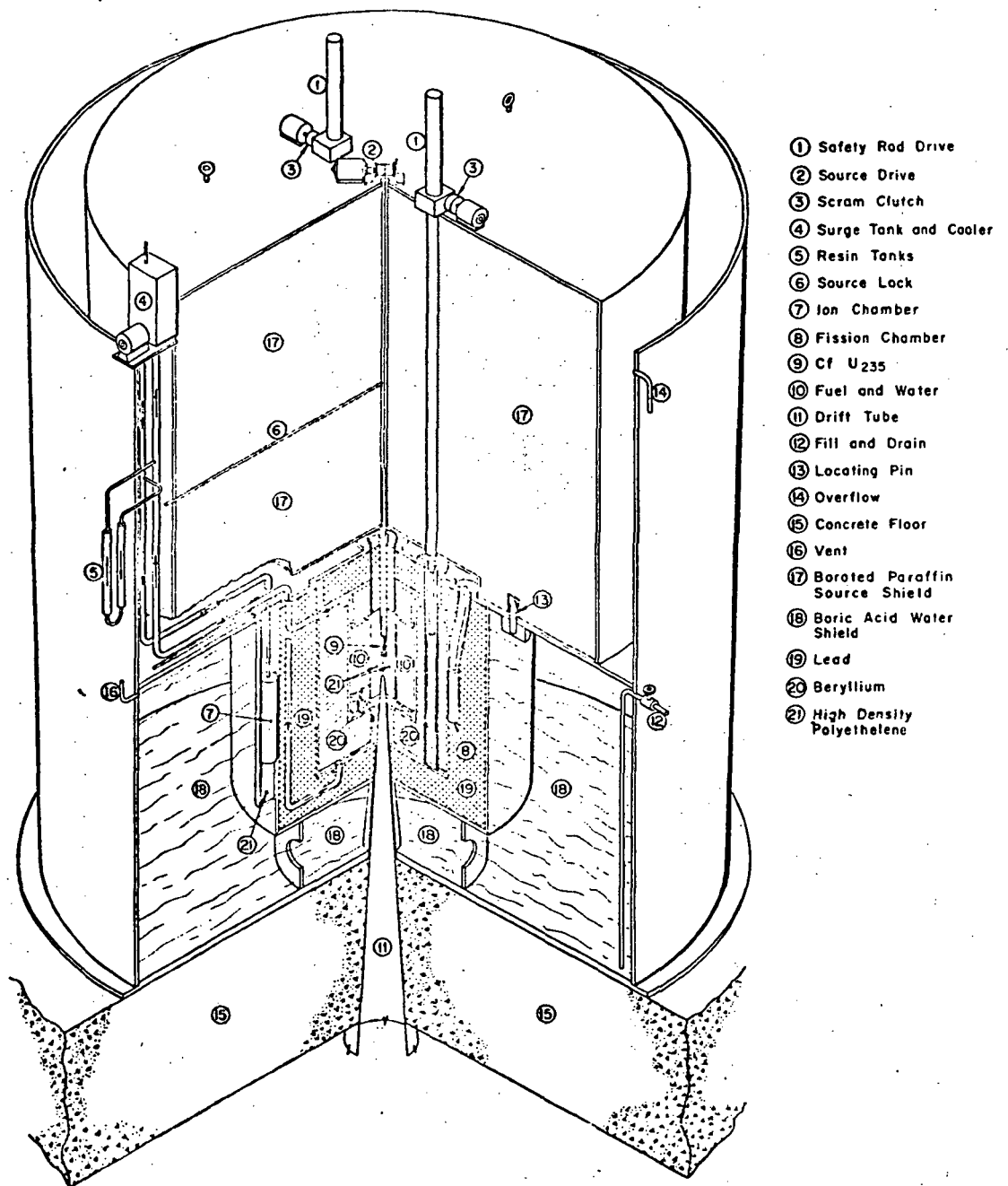
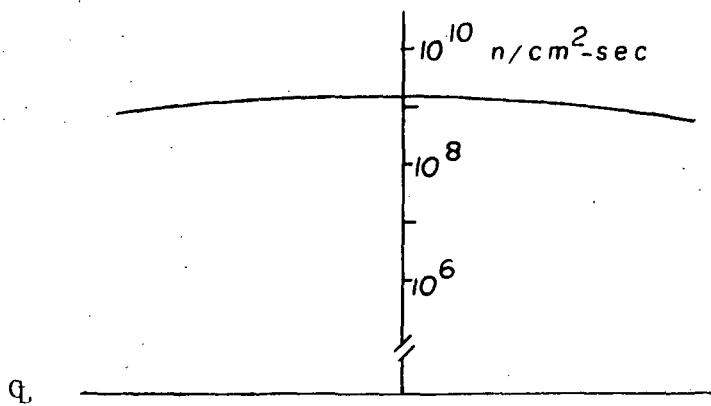
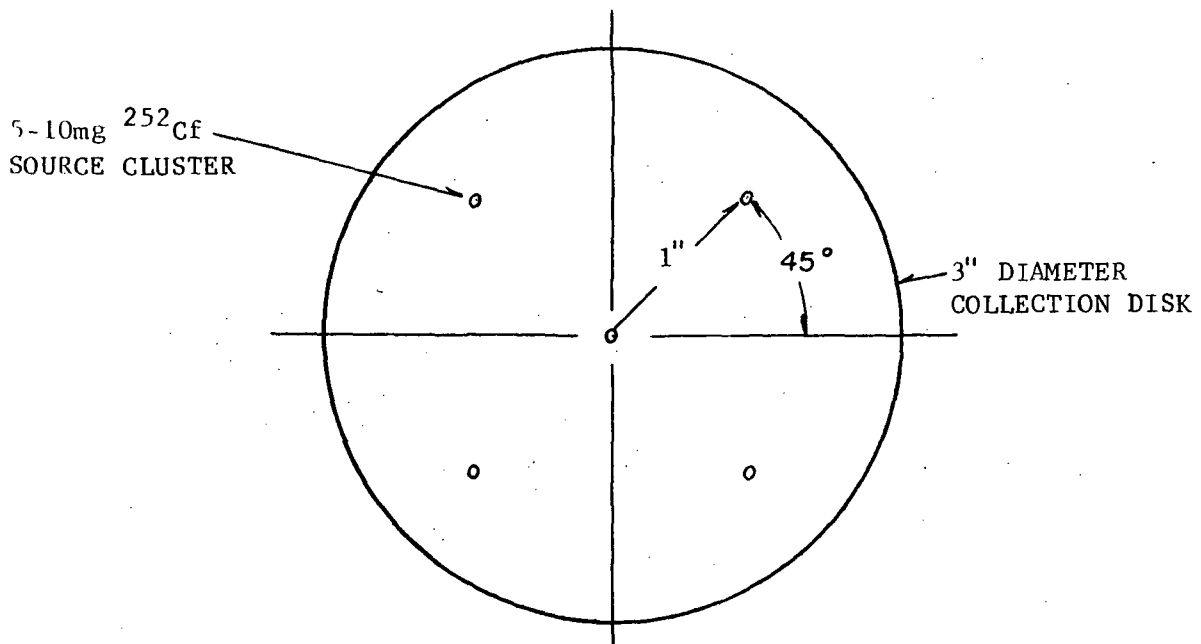


Fig. 3-2. Cutaway view of a subcritical  $^{252}\text{Cf}$  neutron multiplier.  
Upper section shields the source when not in use.  
(Based on G-16)

Fig. 3-3. SOURCE MATRIX DESIGN



NEUTRON FLUX PROFILE

irradiation.

Since the portability of any isotopic neutron source is only limited by its shielding requirements, a comprehensive shielding study was made. To obtain reliable estimates the neutron source configuration was assumed to be a point source of 50 milligrams. The radiations emitted by a Cf-252 source are listed and evaluated under the four groupings below.

Fast neutrons: Most of the fast neutrons result from the spontaneous fission of Cf-252 with lesser amounts coming from ( $\alpha, n$ ) reactions on low atomic number elements. These fast neutrons are distributed according to a typical fission spectrum up to 13 MeV in energy. The most effective shielding of fast neutrons is a hydrogenous mixture.

Thermal neutrons: Thermal neutrons result primarily from the slowing down of the fast neutrons. The most effective shield in this case is a material with a high thermal cross-section such as boron or lithium.

Primary gammas: The primary gamma rays are a result of spontaneous fission, alpha decay of Cf-252 and fission product decay. The most effective shielding for this type of radiation is a high density material such as

iron or lead.

Secondary gammas: Secondary gammas are created as a result of the capture of thermal neutrons by hydrogen nuclei. This radiation can be minimized by adding boron or lithium to the hydrogenous shield.

In arriving at a optimal shielding design, several generalizations and assumptions were made:

1. Thermal neutrons will contribute a relatively small portion of the overall dose.
2. The primary gamma radiation may be eliminated by relatively small amounts of lead or iron.
3. The fast neutron and secondary gamma provide the major portion of the radiation dose at the outside of the shield.
4. The Cf-252 source is dispersed in a 1 cm<sup>3</sup> aluminum capsule at the center of the shielding.
5. The shielding can be protected against chamber environment.

From the above conditions and assumptions, it is concluded that the most effective shielding will consist of a borated polyethylene and lead composition. Several comparative shielding design proposals are presented in Table 3-9. These estimates are based on data from AEC Research and Development Report (G-15). These shielding designs offer a combined neutron and gamma dose rate at the shield surface of less than 10 mrem/hr or

Table 3-9. SHIELDING DESIGN

SHIELD DESIGN	TOTAL DOSE RATE (mrem/hr)	WEIGHT (1000 #)	VOLUME (ft <sup>3</sup> )
33" PARAFFIN, 2" LEAD, 2 mg/ccB	10.4	4.7	25
33" PARAFFIN, 2.25" LEAD, 2 mg/ccB	8.9	4.76	25.3
33" POLYETHYLENE, 1.75" LEAD, 2 mg/ccB	8.5	4.6	24.7
40" PARAFFIN, 3" LEAD, 2 mg/ccB	.8	14.8	61.2

(a) Radial Dimensions  
 (b) 6" x 6" Source Opening Considered  
 (c) For 50 mg point source of Cf-252

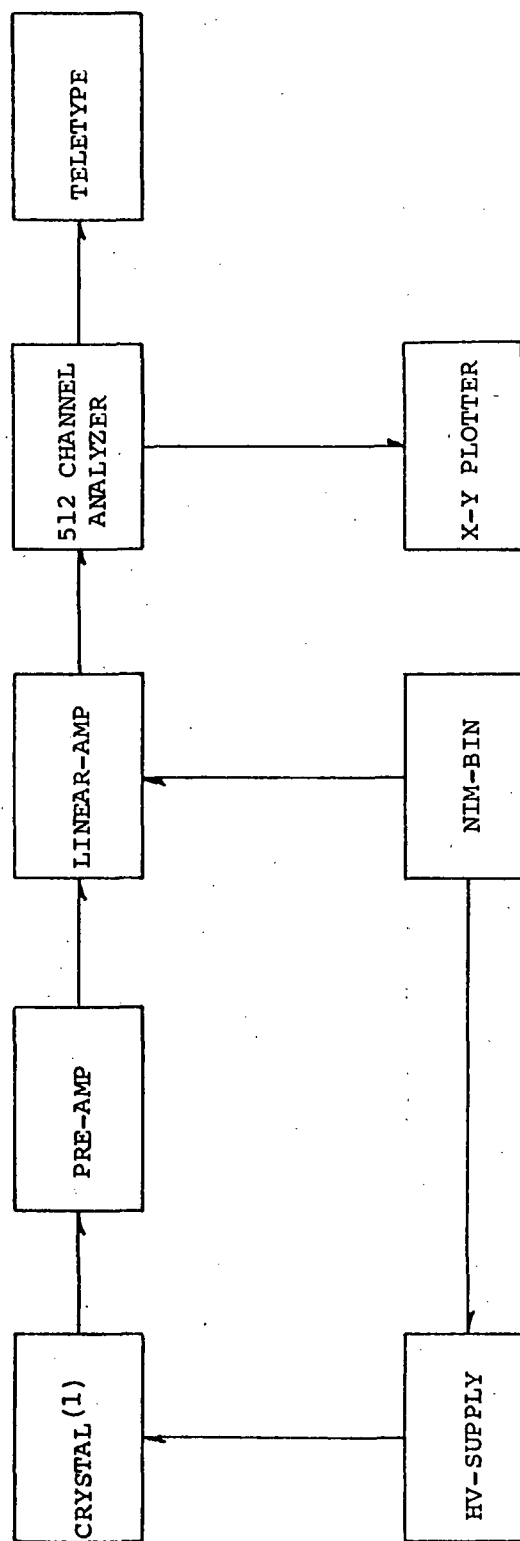
1 mrem/hr. The weight and volume information was obtained by considering a spherical shield design which results in minimum system weight. Using the 0.8 mrem/hr surface dose rate as a design criteria, the area outside the chamber would satisfy radiation safety requirements for an uncontrolled area with a calculated dose rate level of 0.21 mrem/hr. Shielding calculations are given in Appendix B.

A brief description of the instrumentation and electronics of the proposed detection system will be provided to complete the system description. The main component of the radiation counting system is a 512 channel pulse height analyzer coupled to a 3 in. x 3 in. NaI(Tl) scintillation detector. Gamma ray spectra will be read out from the analyzer via a printer and x-y plotter. A block diagram of this gamma ray spectrometer analyzer system is shown in Figure 3-4.

An estimate of the cost for the Cf-252 on-line monitoring system is given in Table 3-10. The total of \$525,000 includes all system components but does not include the additional costs associated with tagging potential contaminant materials. It should be pointed out that the \$500,000 cost figure for 50 mg of Cf-252 is based on the current AEC price of \$10 per microgram,



Fig. 3-4. GAMMA RAY SPECTROMETRY SYSTEM (BLOCK DIAGRAM)



(1) NaI(Tl) Crystal, Phototube and Voltage Divider

Table 3-10. <sup>252</sup>Cf ON-LINE SYSTEM PRICE LIST

<u>Manufacturer</u>	<u>Type</u>	<u>Name</u>	<u>Price (Dollars)</u>
USAEC Savannah River Laboratories		50 mg <sup>252</sup> Cf	500,000
USAEC Savannah River Laboratories		Cf Packing & Handling	8,000
Harshaw Chemical Co.	12S12L	3x3 NaI(Tl) Crystal, Phototube	850
Tennelec, Inc.	TB-3/TC911	Power bin (NIM type)	526
Tennelec, Inc.	TC940	High Voltage Supply	426
Tennelec, Inc.	TC155A	Pre-Amp	151
Tennelec, Inc.	TC202BLR	Linear-Amp	401
Northern Scientific, Inc.	NS-102	Teletype Series 33	1,050
Northern Scientific, Inc.	NS-606	512 Channel Analyzer	4,950
Hewlett Packard	7034A	X-Y Recorder	1,195
Reactor Experi- ments, Inc.		Shielding	1,050
N/A		Fabrication Costs	6,401
Total			<hr/> \$525,000

- (a) Estimated cost in dollars  
(b) Prices subject to change

Note: This is not a recommendation of any specific manufacturer or an endorsement of his products but is intended for information purposes only.

which has been established from a rather short production and sales history. As a government agency, NASA may be able to negotiate with the AEC to obtain a more favorable pricing arrangement for a 50 mg source. If AEC projections of Cf-252 price for the mid-1970's is accurate, a total system cost of approximately \$125,000 could be expected (based on Cf-252 costs of \$2 per microgram). Table 3-6 shows a cost comparison of this system with three alternative approaches.

## 4.0 CONCLUSIONS AND RECOMMENDATIONS

### 4.1 CONCLUSIONS

The methods which have been considered and their characteristics are reviewed in Table 4-1. Of those considered, only the alpha range technique has the required sensitivity and meets the radiological safety requirements. However, this technique will not determine the source of the contamination, but like the quartz microbalance presently in use, functions only as a gross contamination detector. It does have the unique capability of detecting noncondensable contaminants in the chamber atmosphere. These would seem to be of less importance than condensable ones, but when used in conjunction with a quartz microbalance, the two systems would yield total and condensable contaminant levels from noncondensable levels could be estimated.

The strong directional dependence of the alpha range system may be either an asset or a liability depending on the use envisioned for the system. This dependence should be kept in mind as well as the requirement that the detector track over a considerable length of the tubes

Table 4-1. RADIOISOTOPE METHODS SUMMARY

Method	Detectable Limit g/cm <sup>2</sup>	Radiological Safety Status	Advantages	Disadvantages	Cost
<sup>14</sup> C tag	$1.3 \times 10^{-4}$	Unacceptable	Selective testing of suspect agents on an individual basis. Operational over complete cycle.	Long half life isotope. Tag may not follow volatile component.	Not computed due to unacceptable radiological safety status.
<sup>3</sup> H tag	$2.7 \times 10^{-7}$	Unacceptable	Selective testing of suspect agents on an individual basis. Operational over complete cycle.	Long half life isotope. Tag may not follow volatile component.	Not computed due to unacceptable radiological safety status.
Trace Element Tag	$2.9 \times 10^{-4}$	Acceptable	Selective testing of suspect agents on an individual basis. Operational over complete cycle.	Tag may not follow volatile component.	Not computed due to unacceptable sensitivity capability.
Alpha Range	$1.9 \times 10^{-10}$	Acceptable	Operational over complete cycle. Detects both condensable and non-condensable contaminants. Directional dependent. Gas phase sensitivity of $2 \times 10^{-13}$ g/cm <sup>3</sup> .	Size problems, 10 meters long. Sensitivity directional dependent. New system never used before for this purpose.	\$37,000 - \$50,000 for equipment.
Fission Product Range	$2.3 \times 10^{-6}$	Questionable	Same as those of alpha range.	Same as those of alpha range. Lower sensitivity than alpha range.	In excess of \$50,000 for equipment

during the pump down and repressurization stages of the chamber cycle.

The equipment selected for use in the alpha system is notable only in that the detector selected is a ruggedized light tight version with an aluminum plated window. Thus, chamber light conditions will have no effect on the detectors.

The tritium method is the most sensitive of the tagging techniques and is suggested for testing of suspect agents in a small chamber dedicated to this purpose. The ability to tag a suspect agent will depend upon its chemical compound; however, the  $7 \times 10^{-8}\%$  tag is thought to be a conservative estimate.

While neutron activation analysis is well known as a highly sensitive trace analytical technique, it must also be recognized that ultimate sensitivities are achievable only under favorable conditions. This study has shown that conditions are very unfavorable for the on-site detection and identification of microgram quantities of space environmental chamber contaminants by neutron activation analysis techniques as summarized in Table 4-2. Additional factors supporting this conclusion are given below:

Lack of Inherent Contaminant "Fingerprint": Contaminant materials studied are composed of elements which

Table 4-2. NEUTRON ACTIVATION ANALYSIS METHODS SUMMARY

Technique	Initial Cost Analysis (\$)	Analysis Speed	Measurement Sensitivity
On-line $^{252}\text{Cf}$ system (in-chamber)	525,000 (2)	Hourly	Poor
$^{252}\text{Cf}$ and Multiplier system (1) (on-site)	200,000 (2,3)	Daily	Fair
14 MeV generator (on-site)	25,000	Daily	Very Poor
Reactor at Texas A&M University	600 min. (4)	Several Days	Good

(1) 5-3 mg sources with a 4 times multiplier.

(2) Based on \$10 per microgram.

(3) Cost of multiplier approximately \$50,000.

(4) \$300/day for reactor time plus \$30/sample for analysis.  
Minimum 10 samples.

do not have suitable properties for neutron activation. Experimental results indicated no unique neutron activation products useful for contaminant identification.

Contaminant Tagging Stability: Evaluation of chemical stability of proposed tagged compounds under high vacuum and temperature conditions would be required.

Neutron Source Replacement Cost: Cf-252 (2.6 year half-life) would require periodic replenishment at an average annual cost of approximately 20 to 25% of the original source cost.

Reliability Factor: Under "best case" conditions, the expected performance of the in-chamber system can be rated only marginal (Table 3-5). Since the probability of achieving "best case" conditions in actual test operations is unlikely, the in-chamber system would not be a reliable solution.

#### 4.2 RECOMMENDATIONS

The alpha range system is the only practical radioisotope technique with sufficient sensitivity to detect the impurity levels presently being measured in the space simulation chambers. Should it be desirable to independently confirm the quartz microbalance results or determine the gas phase contaminant level, it is recommended that this system be given serious consideration.

In view of the marginal measurement sensitivity achievable with an on-site isotopic neutron activation analysis system, the high cost of the neutron source and the requirement for further research on the chemistry of tagging potential contaminant materials, it is clear that



this approach to contamination identification and monitoring in the NASA space environmental simulation chambers is not feasible.

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APPENDIX B: SAMPLE CALCULATIONS

## APPENDIX B: SAMPLE CALCULATIONS

### TAG LEVEL: (BROMINE)

First, one must determine the average percent by weight of bromine that can be placed in the oils as tags. (Only valid if assumptions 1-4, page 34 are good).

For DC-11, MW = 546.0

By replacing 5Br with 5H, the MW becomes 941 of which a MW of 400 is associated with Bromine.

Therefore,  $\frac{400 \text{ MW}}{941 \text{ MW}_t} = .42$  or 42% Br in DC-11.

Similar calculations involving the other oils will also produce an average concentration around 38%.

Assuming average contamination of  $10^{-6}$  gm/cm<sup>2</sup> (J-1) and a collection disk area of 45.58 cm<sup>2</sup>, the tag limit can be defined as:

$$\text{C. L.} = \frac{(\text{Fraction Br in Contaminant}) (\text{Avg. Contaminant})}{(\text{Area})}$$

$$\text{That is, C. L.} = (.38) (10^{-6} \text{ gm/cm}^2) (45.58 \text{ cm}^2) = 17.29 \times 10^{-6} \text{ gm.}$$

## APPENDIX B: SAMPLE CALCULATIONS

### DETECTION LIMIT: (BROMINE)

$$D. L. = PA/YIELD$$

where PA = Minimal peak area of tag element, and  
YIELD = Tag element yield in cpm/gm.

Background Definition obtained from Appendix C:  
+10% variance from .62 MeV gamma is equal to 13 channels  
Therefore, background average over 13 channel interval  
is approximately 103.9 counts/channel/hour.

$$PA = 2 (BASE)^{1/2}$$

$$\text{where } BASE = \frac{(103.9 \text{ c/ch/hr}) (13 \text{ ch})}{60 \text{ min/hr}} = 22.51 \text{ cpm.}$$

$$\text{Therefore, } PA = 9.48 \text{ cpm.}$$

To determine YIELD, tabular values of photopeak yield  
(in cpm/gm) for a thermal flux of  $4.3 \times 10^{12}$  n/sec/cm<sup>2</sup>  
(I-14) were corrected to values of  $1.25 \times 10^9$  n/sec/cm<sup>2</sup>;  
the thermal flux from a 50 mg point source of Cf-252.

$$\text{Correction Factor} = \frac{1.25 \times 10^9 \text{ n/sec/cm}^2}{4.3 \times 10^{12} \text{ n/sec/cm}^2} = 2.9 \times 10^{-4}$$

Therefore, the corrected photopeak yield for Bromine is,

$$(1.4 \times 10^{10} \text{ cpm/gm}) (2.9 \times 10^{-4}) = 4.06 \times 10^6 \text{ cpm/gm.}$$

Thus, the detection limit is,

$$D. L. = PA/YIELD = \frac{9.48 \text{ cpm}}{4.06 \times 10^6 \text{ cpm/gm}} = 2.3 \times 10^{-6} \text{ gm.}$$

## APPENDIX B: SAMPLE CALCULATIONS

### RELIABILITY FACTOR: (BROMINE)

$$R = \frac{(\%Tag) (Den) (Area)}{(Det)}$$

where,

%Tag = Maximum % tag element in contaminants

Den = Average contamination density on collection disk

Det = Detection limit of the tag element in  $\mu\text{gms.}$

Area = Area of 3 inch diameter collection disk  
(45.58  $\text{cm}^2$ )

Therefore,

$$R = \frac{\text{Contamination Limit}}{\text{Detection Limit}}$$

$$R = \frac{17.29 \times 10^{-6} \text{gm}}{2.3 \times 10^{-6} \text{gm}} = 7.4$$



## APPENDIX B: SAMPLE CALCULATIONS

SHIELDING CALCULATIONS: (.8 mrem/hr design)

40" Paraffin, 3" Lead, 2 mg/cc  $^{10}\text{B}$  Composition.  
Source =  $5 \times 10^{-2}$  gms Cf-252.

Estimation from E. I. Du Pont de Nemours & Company by H. E. Hootman (G-15).

Neutron Dose Rate: (From figure 2)

NDR = .5 mrem/hr

This is the neutron dose rate at the surface of the shield. To obtain the neutron dose rate outside the chamber, the following neutron attenuation formula was used:

$$I = I_0 e^{-\sigma_n x}$$

where,

$$\begin{aligned} \sigma_n &= \rho N_O / A = \frac{(7.83 \text{ gm/cc}) (6.02 \times 10^{23} \text{ mol}^{-1}) (2.5 \text{ b})}{26} \\ &= .45 \text{ cm}^{-1} \end{aligned}$$

for an estimated chamber wall (steel) thickness of 1 inch.

$$I = I_0 (.31) = (.5 \text{ mrem/hr}) (.31) = .158 \text{ mrem/hr.}$$

Total Gamma Dose Rate: (from Fig. 3, 5, & 7 respectively)  
 $\text{GDR} = (8 \times 10^{-3}) (10^{-2}) (7.5 \times 10^{-2}) = 6 \times 10^{-6} \text{ mrem/hr/}\mu\text{gm}$ , or .3 mrem/hr

This is the total gamma dose rate at the surface of the proposed shield. Using a similar analysis, the total dose rate outside the chamber can be obtained using the following formula:

$$I = I_0 e^{-\mu x}$$

where,  $\mu$  is the linear absorption coefficient. The linear absorption coefficient was found to be  $.72 \text{ cm}^{-1}$  (using the average secondary gamma ray to be .48 MeV).

Therefore,

$$I = I_0 (.16) = .3 \text{ mrem/hr} (.16) = 0.48 \text{ mrem/hr}$$

SHIELDING CALCULATIONS: (CONTINUED)

The combined total gamma and neutron dose rate is .158 mrem/hr plus .048 mrem/hr which equals .206 mrem/hr.

This quantity is less than the I. C. R. P. recommendation of .25mr/hr.\* for the population at large.

\*Rees, D. J., Health Physics, MIT Press, 1967, pp. 130.

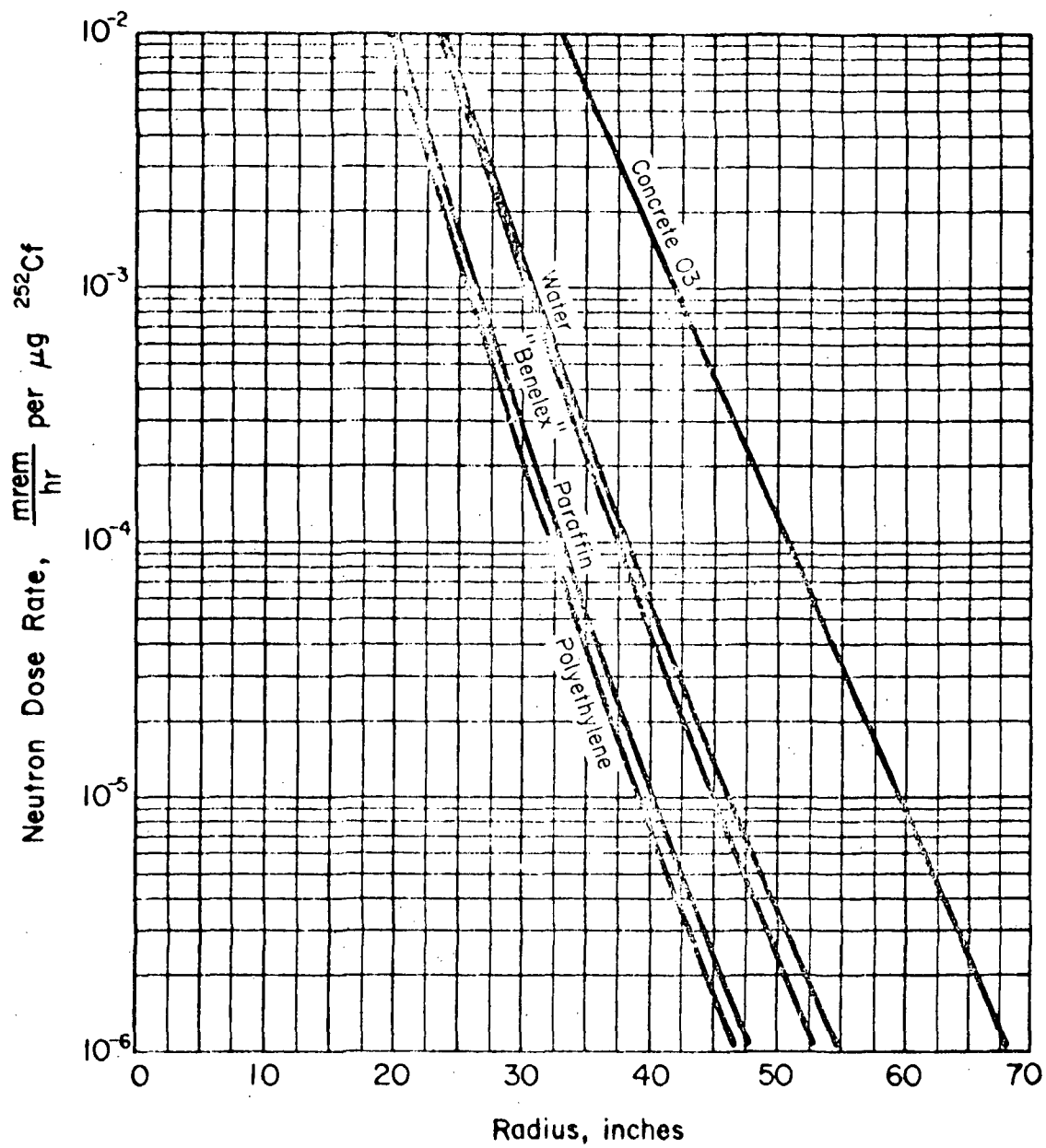


FIG. 2 (Continued)

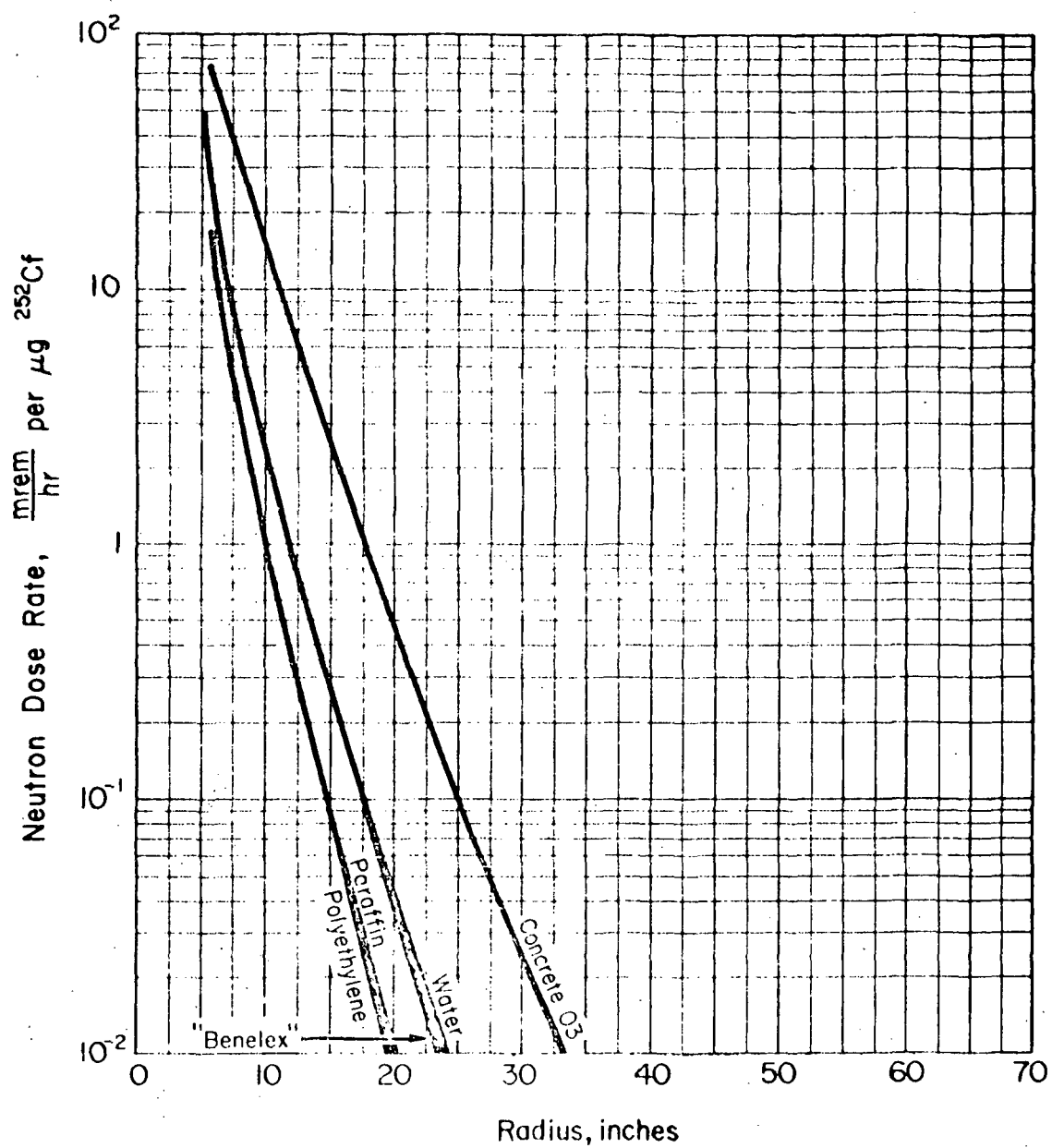


FIG. 2 NEUTRON DOSE RATE

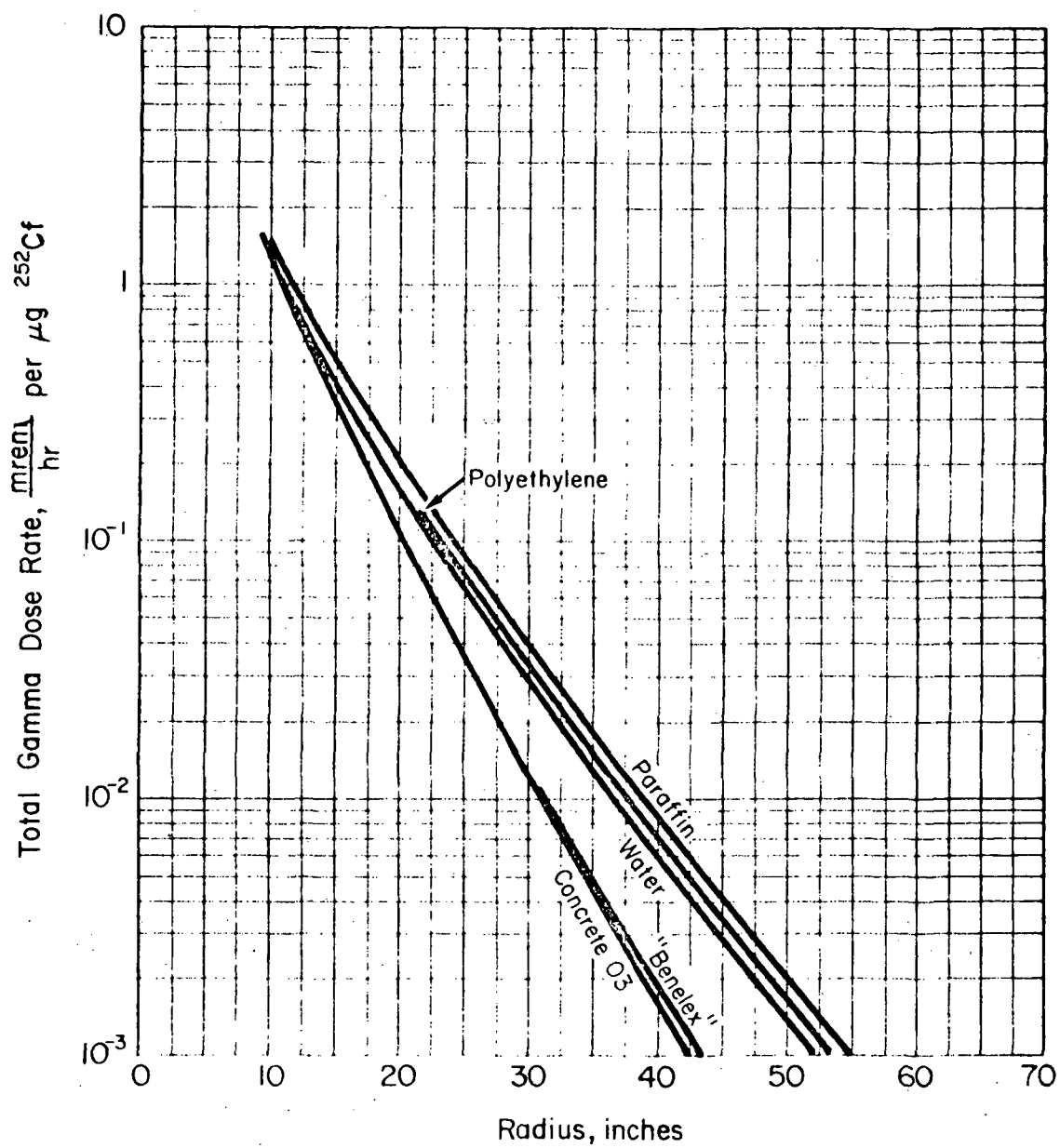


FIG. 3 TOTAL GAMMA DOSE RATE

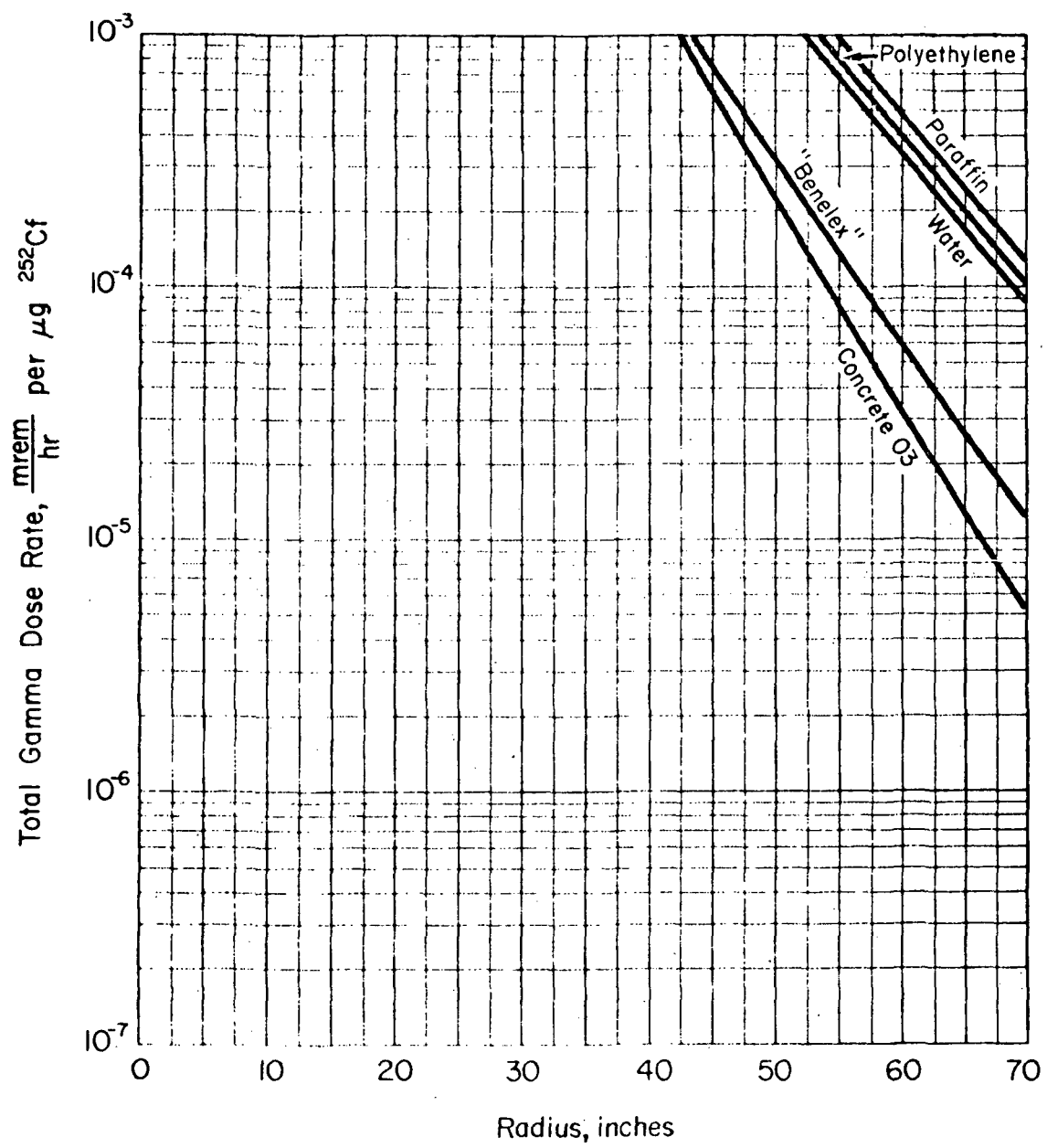


FIG. 3 (Continued)

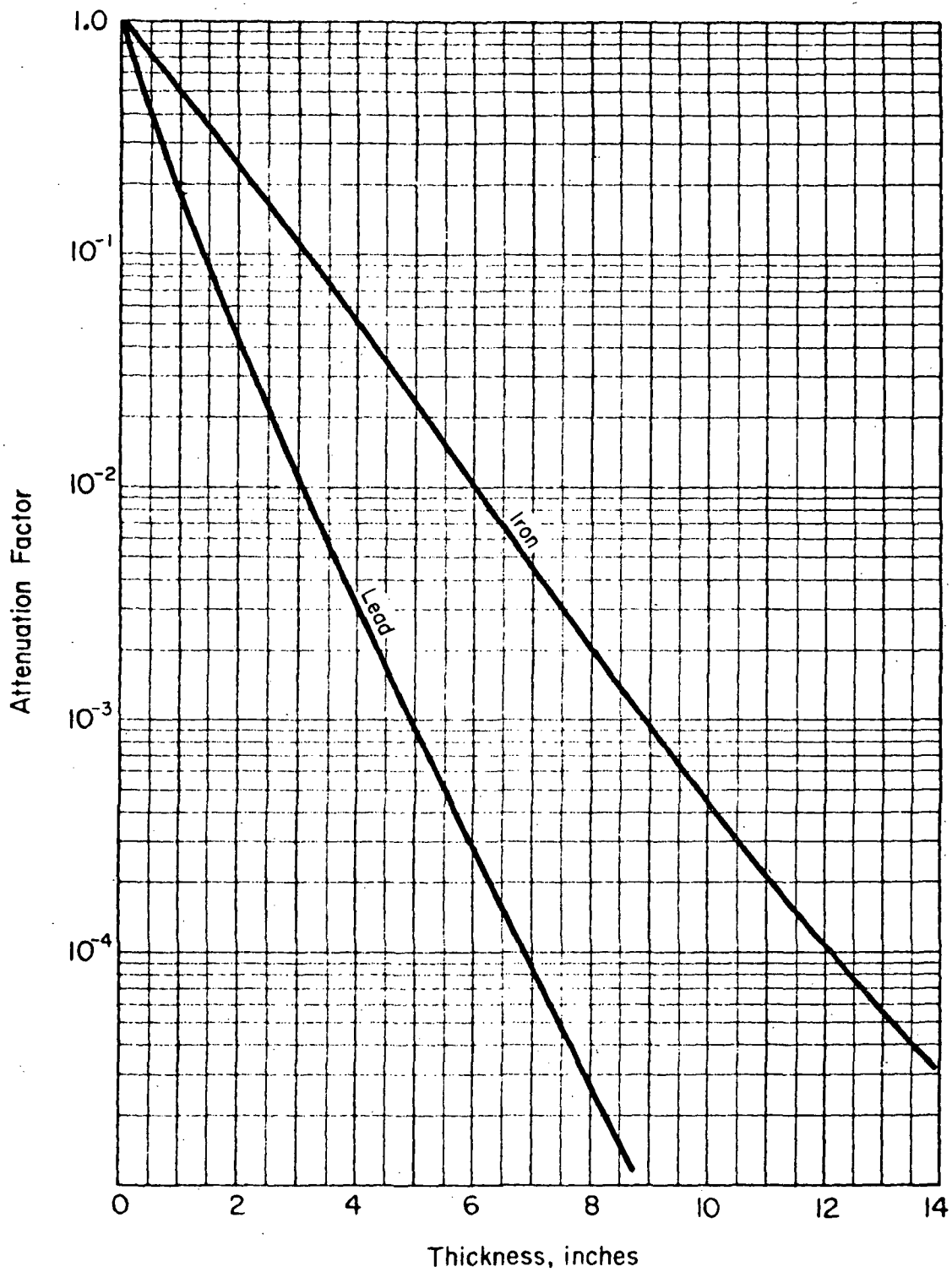


FIG. 5 ATTENUATION OF PROMPT FISSION GAMMA RAYS  
FROM A POINT ISOTROPIC SOURCE OF  $^{252}\text{Cf}$

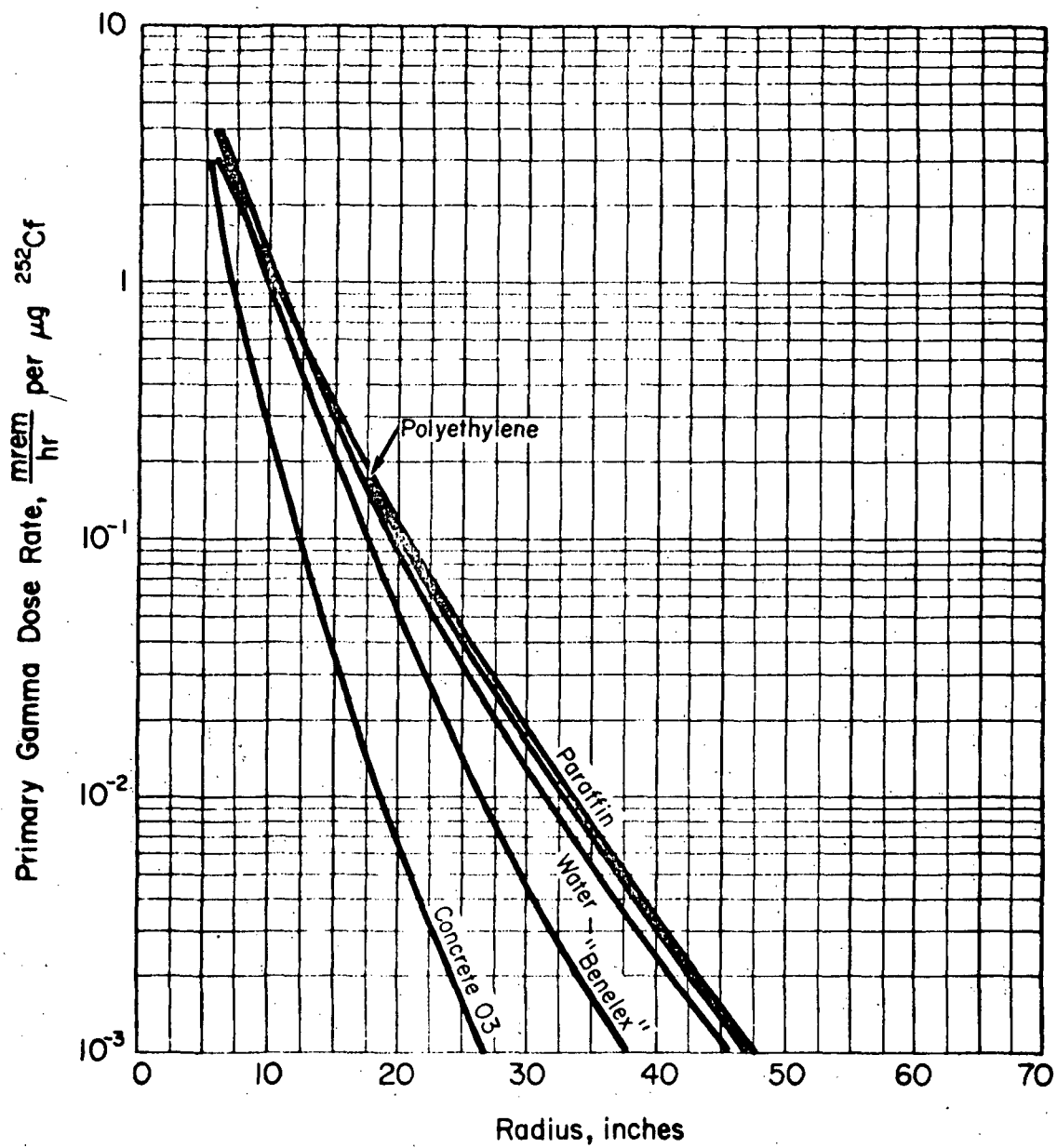


FIG. 6 PRIMARY GAMMA DOSE RATE



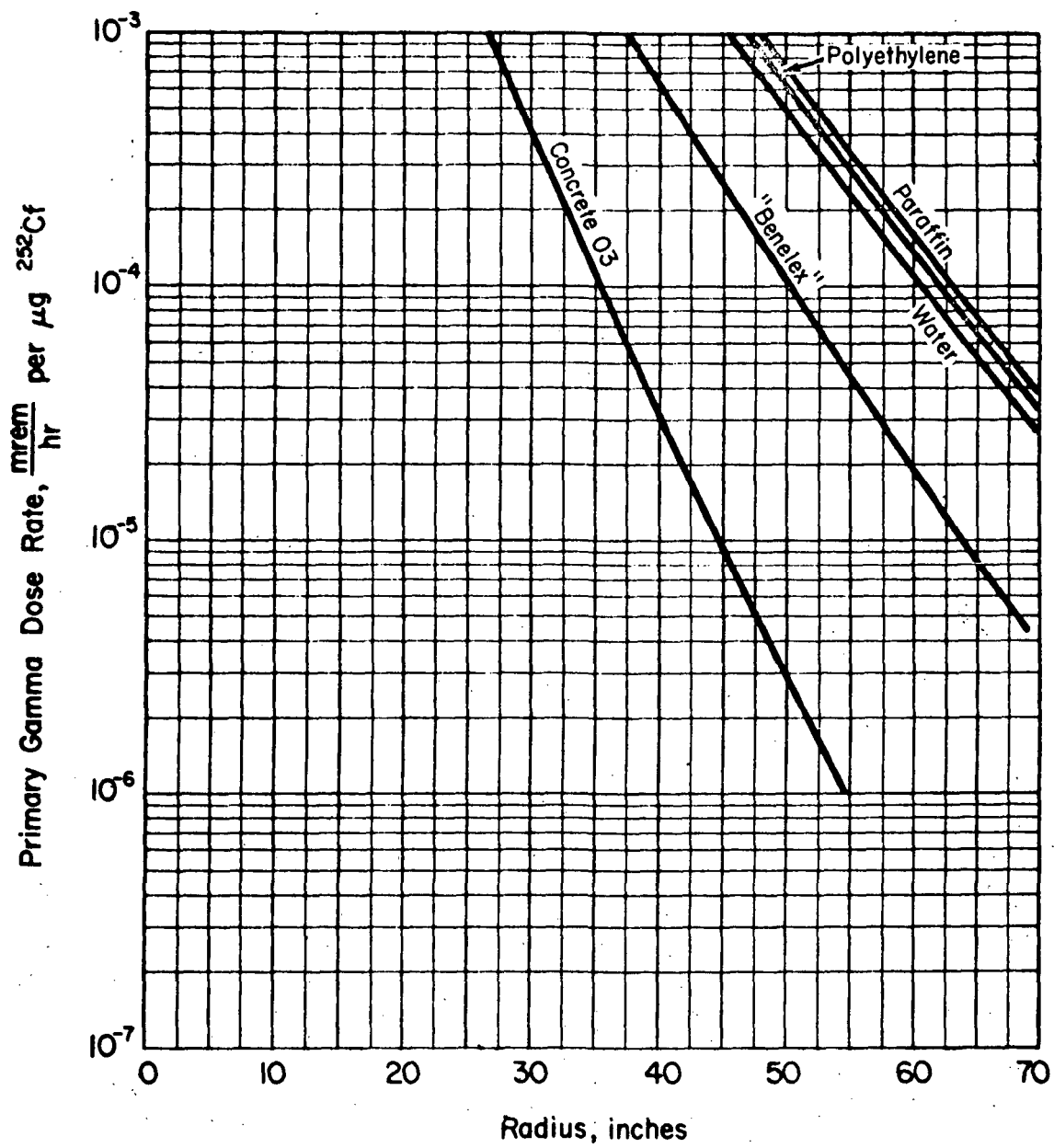


FIG. 6 (Continued)

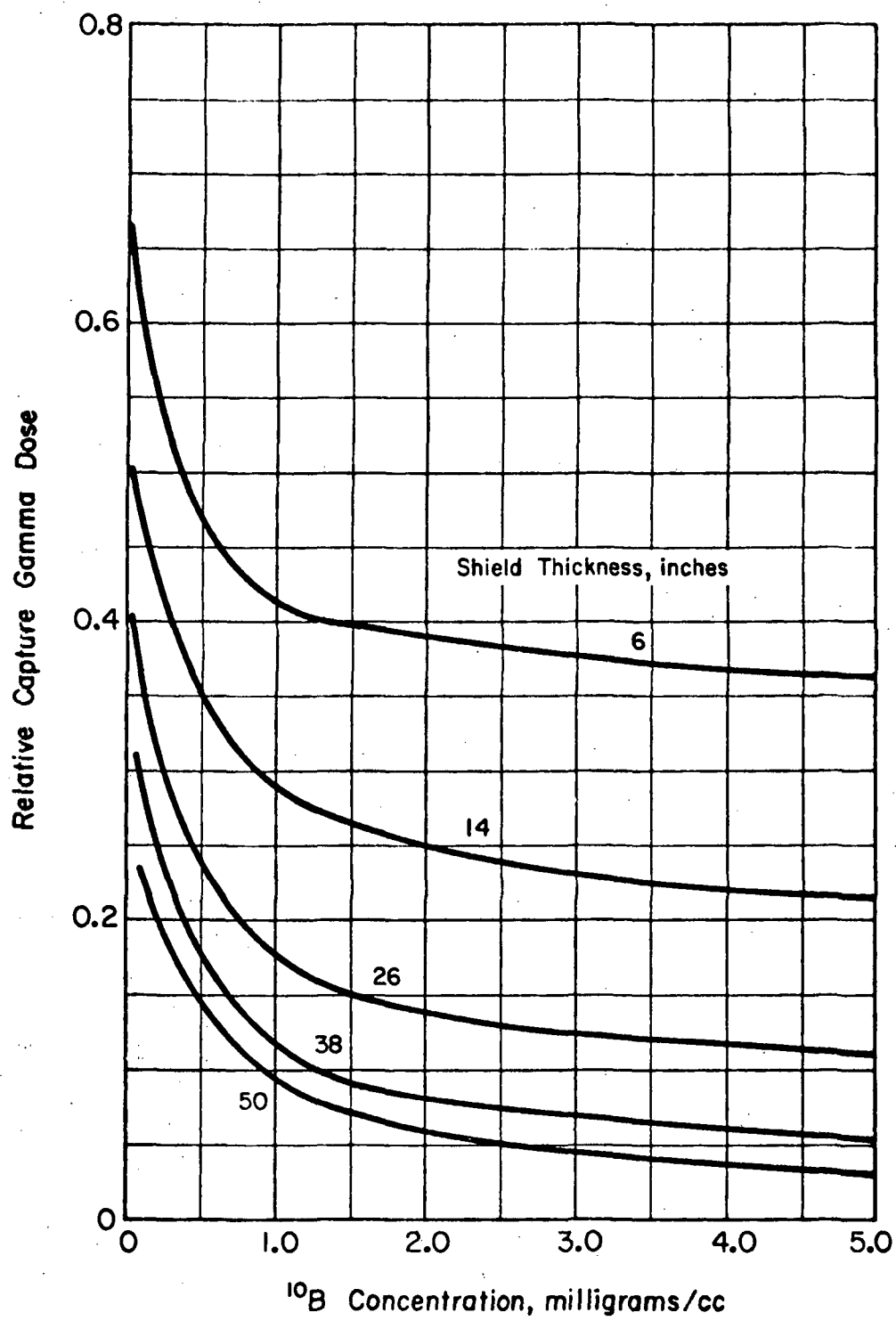


FIG. 7 EFFECT OF BORATION ON RELATIVE CAPTURE GAMMA DOSE

# APPENDIX C: BACKGROUND DEFINITION

Background is defined as the gamma ray spectra obtained from a one hour count on a shielded NaI 400 channel gamma ray spectrometer. The bracketed sections indicate the background in that region of the spectrum associated with each tag element.

	1- 40	41- 80	81- 120	121- 160	161- 200
1	0.	147.-	74.+	40.-	39.-
2	0.	135.-	65.-	41.+	23.-
3	313.+	123.-	80.-	45.+	36.+
4	215.+	138.-	70.-	47.+	36.+
5	185.-	122.+	69.-	37.2	25.+
6	144.-	130.+	66.-	46.+	39.+
7	129.-	131.+	71.+	46.-	33.+
8	142.+	141.+	60.+	42.-	37.+
9	174.+	146.+	67.+	39.+	38.+
10	159.+	165.+	78.+	48.+	38.-
11	154.+	174.-	73.-	40.+	45.+
12	180.+	168.-	74.-	40.+	29.+
13	177.+	123.-	71.-	46.+	33.+
14	201.+	106.-	57.-	50.+	40.+
15	224.+	127.+	53.-	52.+	42.+
16	237.+	127.+	60.-	45.+	41.+
17	265.+	136.-	71.+	49.+	35.+
18	257.+	119.-	55.+	49.+	38.-
19	256.+	124.-	61.-	67.+	39.-
20	276.+	113.-	59.-	52.+	40.+
21	298.+	112.-	74.-	46.-	39.+
22	260.-	101.-	43.-	58.-	36.+
23	278.-	85.-	54.-	48.-	40.+
24	235.-	94.-	58.-	49.-	47.+
25	245.-	96.-	62.+	39.-	33.+
26	244.-	81.-	58.+	38.-	39.+
27	250.-	82.-	44.-	39.-	48.+
28	209.-	86.-	57.-	38.-	49.+
29	209.-	72.-	61.+	32.-	50.+
30	193.-	82.-	56.+	32.-	48.+
31	202.-	75.-	55.-	32.-	47.-
32	195.-	84.-	62.-	31.-	41.+
33	170.-	63.-	59.-	23.-	54.+
34	194.-	63.-	51.-	24.+	49.+
35	167.-	64.-	50.-	36.+	41.+
36	155.-	77.+	59.-	32.+	56.+
37	165.-	74.+	49.-	28.+	54.+
38	153.-	67.+	42.-	45.-	47.+
39	155.-	70.-	31.-	29.-	47.+
40	140.-	73.+	42.-	23.+	72.+

# **APPENDIX D** **DOT REGULATIONS**

Excerpts from

*Tariff No. 23 - Hazardous Materials Regulations of  
the Department of Transportation*

Brackets apply specifically to californium.

§ 173.389 Radioactive materials; definitions. For the purpose of Parts 170-159:

(a) "Fissile radioactive material" means the following material: Plutonium-238, plutonium-239, plutonium-241, uranium-233, or uranium-235, or any material containing any of the foregoing materials. See § 173.396 (a) for exclusions. Fissile radioactive material packages are classified according to the controls needed to provide nuclear criticality safety during transportation as follows:

(1) Fissile Class I. Packages which may be transported in unlimited numbers and in any arrangement, and which require no nuclear criticality safety controls during transportation. For purposes of nuclear criticality safety control, a transport index is not assigned to Fissile Class I packages. However, the external radiation levels may require a transport index number.

(2) Fissile Class II. Packages which may be transported together in any arrangement but in numbers which do not exceed an aggregate transport index of 50. For purposes of nuclear criticality safety control, individual packages may have a transport index of not less than 0.1 and not more than 10. However, the external radiation levels may require a higher transport index number but not to exceed 10. Such shipments require no nuclear criticality safety control by the shipper during transportation.

(3) Fissile Class III. Shipments of packages which do not meet the requirements of Fissile Class I or II and which are controlled to provide nuclear criticality safety in transportation by special arrangements between the shipper and the carrier.

Note 1: Uranium-235 exists only in combination with various percentages of uranium-238 and uranium-239. "Fissile radioactive material" as applied to uranium-235 refers to the amount of uranium-235 actually contained in the total quantity of uranium being transported.

Note 2: Radioactive material may consist of mixtures of fissile and non-fissile radionuclides. "Fissile radioactive material" refers to the amount of plutonium-238, plutonium-239, plutonium-241, uranium-233, uranium-235, or any combination thereof actually contained in the mixture. The "radioactivity" of the mixture consists of the total activity of both the fissile and nonfissile radionuclides. All mixtures containing "fissile material" shall be subject to § 173.395.

(b) "Large quantity radioactive materials" means a quantity the aggregate radioactivity of which exceeds that specified as follows:

(1) Groups I or II (see paragraph (h) of this section) radionuclides: 20 curies.

(2) Groups III or IV radionuclides: 200 curies.

(3) Group V radionuclides: 5,000 curies.

(4) Groups VI or VII radionuclides: 50,000 curies.

(5) Special form material: 5,000 curies.

(c) "Low specific activity material" means any of the following:

(1) Uranium or thorium ores and physical or chemical concentrates of those ores;

(2) Unirradiated natural or depleted uranium or unirradiated natural thorium;

(3) Tritium oxide in aqueous solutions provided the concentration does not exceed 5 millicuries per milliliter;

(4) Material in which the activity is essentially uniformly distributed and in which the estimated average concentration per gram of contents does not exceed:

(i) 0.0001 millicuries of Group I (see § 173.389 (h)) radionuclides; or

(ii) 0.005 millicuries of Group II radionuclides; or

(iii) 0.3 millicuries of Groups III or IV radionuclides.

Note: This includes, but is not limited to, materials of low radioactivity concentration such as residues or solutions from chemical processing, wastes such as building rubble, metal, wood, and fabric scrap, glassware, paper and cardboard, solid or liquid plant waste, sludges, and ashes.

(5) Objects of non-radioactive material externally contaminated with radioactive material, provided that the radioactive material is not readily dispersible and the surface contamination when averaged over an area of 1 square meter, does not exceed 0.0001 millicurie (220,000 disintegrations per minute) per square centimeter of Group I radionuclides or 0.001 millicurie (2,200,000 disintegrations per minute) per square centimeter of other radionuclides.

(d) "Normal form radioactive materials" means those which are not special form radioactive materials. Normal form radioactive materials are grouped into transport groups (see paragraph (h) of this section).

(e) "Radioactive material" means any material or combination of materials, which spontaneously emits ionizing radiation. Materials in which the estimated specific activity is not greater than 0.002 microcuries per gram of material, and in which the radioactivity is essentially uniformly distributed, are not considered to be radioactive materials.

(f) "Removable radioactive contamination" means radioactive contamination which can be readily removed in measurable quantities by wiping the contaminated surface with an absorbent material. The measurable quantities shall be considered as being not significant if they do not exceed the limits specified in § 173.397.

(g) "Special form radioactive materials" means those which, if released from a package, might present some direct radiation hazard but would present little hazard due to radiotoxicity and little possibility of contamination. This may be the result of inherent properties of the material (such as metals or alloys), or acquired characteristics, as through encapsulation. The criteria for determining whether a material meets the definition of special form are prescribed in § 173.398 (a).

(h) "Transport group" means any one of seven groups into which normal form radionuclides are classified according to their radiotoxicity and their relative potential hazard in transportation, and as listed in § 173.390.

(i) "Transport index" means the number placed on a package to designate the degree of control to be exercised by the carrier during transportation. The transport index to be assigned to a package of radioactive materials shall be determined by either subparagraph (1) or (2) of this paragraph, whichever is larger. The number expressing the transport index shall be rounded up to the next highest tenth; e.g., 1.01 becomes 1.1.

(1) The highest radiation dose rate, in millirem per hour at three feet from any accessible external surface of the package; or

(2) For Fissile Class II packages only, the transport index number calculated by dividing the number "50" by the number of similar packages which may be transported together (see § 173.396), as determined by the procedures prescribed in the regulations of the U. S. Atomic Energy Commission, Title 10, Code of Federal Regulations, Part 71.

(j) "Type A packaging" means packaging which is designed in accordance with the general packaging requirements of §§ 173.24 and 173.393, and which is adequate to prevent the loss or dispersal of the radioactive contents and to retain the efficiency of its radiation shielding properties if the package is subject to the tests prescribed in § 173.398 (b).

(k) "Type B packaging" means packaging which meets the standards for Type A packaging, and, in addition, meets the standards for hypothetical accident conditions of transportation as prescribed in § 173.398 (c).

(l) "Type A quantity" and "Type B quantity" radioactive materials means a quantity the aggregate radioactivity of which does not exceed that specified as follows:

Transport group (see § 173.389(h))	Type A quantity (in curies)	Type B quantity (in curies)
I.....	0.001	20
II.....	0.03	20
III.....	3	200
IV.....	20	200
V.....	20	1,000
VI and VII.....	1,000	50,000
Special form.....	20	5,000

§ 173.390 Transport groups of radionuclides. (a) List of radionuclides:

Element <sup>1</sup>	Radionuclide <sup>2</sup>	Transport group						
		I	II	III	IV	V	VI	VII
Actinium (89)	Ac-227	X						
	Ac-228	X						
Americium (95)	Am-241	X						
	Am-243	X						
Antimony (51)	Sb-122				X			
	Sb-124			X				
	Sb-125			X				
Argon (18)	Ar-37						X	
	Ar-41							
	Ar-41		X					
	(uncompressed) <sup>3</sup>							
Arsenic (33)	As-73					X		
	As-74					X		
	As-76					X		
	As-77					X		
Astatine (85)	At-211			X				
Barium (56)	Ba-131		X		X			
	Ba-133							
	Ba-140							
Berkelium (97)	Bk-249	X		X				
Beryllium (4)	Be-7					X		
Bismuth (83)	Bi-206					X		
	Bi-207			X				
	Bi-210		X					
	Bi-212			X				
	Bi-214				X			
	Bi-214m				X			
Bromine (35)	Br-82				X			
Cadmium (48)	Cd-109				X			
	Cd-115m			X				
	Cd-115				X			
Calcium (20)	Ca-45				X			
	Ca-47				X			
Californium (98)	Cf-249	X						
	Cf-250	X						
	Cf-252	X						
Carbon (6)	C-14				X			
Cerium (58)	Ce-141				X			
	Ce-143				X			
	Ce-144			X				
Cesium (55)	Cs-131				X			
	Cs-134m			X				
	Cs-134			X				
	Cs-135				X			
	Cs-136				X			
	Cs-137			X				
Chlorine (17)	Cl-36				X			
	Cl-38				X			
Chromium (24)	Cr-51				X			
Cobalt (27)	Co-56			X				
	Co-57				X			
	Co-58m				X			
	Co-58				X			
	Co-60			X				
Copper (29)	Cu-64				X			
Curium (96)	Cm-242	X						
	Cm-243	X						
	Cm-244	X						
	Cm-245	X						
	Cm-246	X						
Dysprosium (66)	Dy-154			X				
	Dy-165				X			
	Dy-166				X			
Erbium (68)	Er-160				X			
	Er-171				X			
Europium (63)	Eu-150				X			
	Eu-152m				X			
	Eu-152			X				
	Eu-154		X					
	Eu-155				X			
Fluorine (9)	F-18				X			
Gadolinium (64)	Gd-153				X			
	Gd-159				X			
Gallium (31)	Ga-67			X				
	Ga-72				X			
Germanium (32)	Ge-71				X			
Gold (79)	Au-193			X				
	Au-194			X				
	Au-195			X				
	Au-196				X			
	Au-198				X			
	Au-199				X			
	Hf-151				X			
	Hf-156				X			
	H-3 (see tritium)							
	Ir-113m				X			
	Ir-114m			X				
	Ir-115m				X			
	Ir-115				X			
	Ir-124			X				
	Ir-125			X				
	Ir-126			X				
	Ir-129			X				
	Ir-131			X				
	Ir-132				X			
	Ir-133			X				
	Ir-134				X			
	Ir-135				X			
	Ir-190				X			
	Ir-192		X					
	Ir-194				X			
	Fe-55				X			
Iron (26)	Fe-50				X			

Element <sup>1</sup>	Radionuclide <sup>2</sup>	Transport group						
		I	II	III	IV	V	VI	VII
Krypton (36)	Kr-80m			X				
	Kr-80m							
	(uncompressed) <sup>3</sup>					X		
	Kr-85			X				
	Kr-85							
(uncompressed) <sup>3</sup>	Kr-87						X	
	Kr-87		X					
	Kr-87							
(uncompressed) <sup>3</sup>	Kr-87					X		
	Kr-87							
Lanthanum (57)	La-140				X			
Lead (82)	Pb-203			X				
	Pb-210							
	Pb-212		X					
Lutecium (71)	Lu-172			X				
	Lu-177				X			
	Lu-177							
Magnesium (12)	Mg-28			X				
Manganese (25)	Mn-52				X			
	Mn-54				X			
	Mn-56				X			
Mercury (80)	Hg-197m							
	Hg-197							
	Hg-203				X			
Mixed Fission Products	Mf-P		X					
	Mf-P							
	Mf-P							
Molybdenum (42)	Mo-99				X			
Neodymium (60)	Nd-147				X			
	Nd-149				X			
	Nd-149							
Neptunium (93)	Np-237	X						
Nickel (28)	Np-239	X						
	Ni-56			X				
	Ni-59				X			
Niobium (41)	Ni-63				X			
	Ni-65				X			
	Nb-94m				X			
Osmium (76)	Nb-95				X			
	Nb-97				X			
	Os-185				X			
Palladium (46)	Os-191m				X			
	Os-191				X			
	Os-193				X			
Phosphorus (15)	Pd-103				X			
	Pd-109				X			
	P-32				X			
Platinum (78)	Pt-191				X			
	Pt-193				X			
	Pt-193m				X			
Plutonium (94)	Pt-197m				X			
	Pt-197							
	Pu-238 <sup>4</sup>	X						
Polonium (84)	Pu-239 <sup>4</sup>	X						
	Pu-240	X						
	Pu-241 <sup>4</sup>	X						
Potassium (19)	Pu-242	X						
	Po-210	X						
	K-42				X			
Praseodymium (59)	K-43			X				
	Pr-142				X			
	Pr-143				X			
Promethium (61)	Pr-143				X			
	Pm-147				X			
	Pm-149				X			
Protactinium (91)	Pm-149				X			
	Pa-231	X						
	Pa-233	X						
Radium (88)	Pa-233		X					
	Ra-223		X					
	Ra-226		X					
Radon (86)	Ra-226				X			
	Ra-228	X						
	Mn-270	X						
Rhenium (75)	Re-222		X					
	Re-183			X				
	Re-186			X				
Rhodium (45)	Re-187			X				
	Re-188			X				
	Re Natural			X				
Rubidium (37)	Rb-103m			X				
	Rb-103			X				
	Rb-86			X				
Ruthenium (44)	Rb-87			X				
	Rb Natural			X				
	Ru-97			X				
Samarium (62)	Ru-103			X				
	Ru-105			X				
	Ru-106			X				
Scandium (21)	Sm-145			X				
	Sm-147			X				
	Sm-151			X				
Selenium (34)	Sm-153			X				
	Sc-46			X				
	Sc-47			X				
Silicon (14)	Sc-48			X				
	Se-75			X				
	Si-31			X				
Silver (47)	Ag-105			X				
	Ag-110m			X				
	Ag-111			X				
Sodium (11)	Na-22			X				
	Na-24			X				
	Sr-83m			X				
Strontium (38)	Sr-83			X				
	Sr-89			X				
	Sr-90		X					
Sulphur (16)	Sr-91			X				
	Sr-92			X				
	S-35			X				
Tantalum (73)	Ta-182			X				
	Tc-96m			X				
	Tc-96			X				
Technetium (43)	Tc-97m			X				
	Tc-97			X				
	Tc-99m			X				
	Tc-99			X				
	Tc-99			X				

Element <sup>1</sup>	Radionuclide <sup>2</sup>	Transport group						
		I	II	III	IV	V	VI	VII
Tellurium (52)	Te-125m				X			
	Te-127m				X			
	Te-127				X			
	Te-129m			X				
	Te-129			X				
	Te-131m			X				
Terbium (65)	Tb-132			X				
	Tb-160			X				
	Tb-200			X				
	Tb-201			X				
Thallium (81)	Tl-202			X				
	Tl-204			X				
	Th-227		X					
	Th-228	X						
Thorium (90)	Tb-230	X						
	Tb-231	X						
	Tb-232			X				
	Tb-234		X					
Thulium (69)	Tb Natural		X					
	Tm-168		X					
	Tm-170		X					
	Tm-171		X					
Tin (50)	Sn-113			X				
	Sn-117m			X				
	Sn-121			X				
	Sn-125			X				
Tritium (1)	H-3			X				
	H-3 (as a gas, as luminous paint, or absorbed on solid materials)			X				X
Tungsten (74)	W-181			X				
	W-185			X				
	W-187			X				
	U-230		X					
Uranium (92)	U-232	X						
	U-233 <sup>3</sup>	X						
	U-234	X						
	U-235 <sup>3</sup>	X						
	U-238	X						
	U Natural	X						
	U Enriched <sup>4</sup>	X						
	U Depleted	X						
Vanadium (23)	V-48			X				
	V-49			X				
	Xe-125			X				
	Xe-131m			X				
Xenon (54)	Xe-131m			X				
	(uncompressed) <sup>5</sup>			X		X		
	Xe-133			X				
	Xe-133 (uncompressed) <sup>5</sup>			X			X	
Ytterbium (70)	Xe-135	X						
	Xe-135 (uncompressed) <sup>5</sup>	X						
	Yb-175			X		X		
	Yb-175			X		X		
Yttrium (39)	Y-88			X				
	Y-90			X				
	Y-91m			X				
	Y-91			X				
Zinc (30)	Y-92			X				
	Y-93			X				
	Zn-65			X				
	Zn-69m			X				
Zirconium (40)	Zr-93			X				
	Zr-95			X				
	Zr-97			X				

<sup>1</sup> Atomic number shown in parentheses.  
<sup>2</sup> Uncompressed means at a pressure not exceeding 14.7 p.s.i. (absolute).  
<sup>3</sup> Atomic weight shown after the radionuclide symbol.  
<sup>4</sup> Fissile radioactive material.

(b) Any radionuclide not listed in the above table shall be assigned to one of the groups in accordance with the following table:

Radionuclide	Radioactive half-life		
	0-1,000 days	1,000 days to 10 <sup>5</sup> years	Over 10 <sup>5</sup> years
Atomic number 1-81	Group III	Group II	Group I
Atomic number 82 and over	Group I	Group I	Group III

NOTE 1: No unlisted radionuclides shall be assigned to Groups IV, V, VI, or VII.

(c) For mixtures of radionuclides the following shall apply:

(1) If the identity and respective activity of each radionuclide are known, the permissible activity of each radionuclide shall be such that the sum, for all groups present, of the ratio between the total activity for each group to the permissible activity for each group will not be greater than unity.

(2) If the groups of the radionuclides are known but the amount in each group cannot be reasonably determined, the mixture shall be assigned to the most restrictive group present.

(3) If the identity of all or some of the radionuclides cannot be reasonably determined, each of those unidentified radionuclides shall be considered as belonging to the most restrictive group which cannot be positively excluded.

(4) Mixtures consisting of a single radioactive decay chain where the radionuclides are in the naturally occurring proportions shall be considered as consisting of a single radionuclide. The group and activity shall be that of the first member present in the chain, except if a radionuclide "x" has a half-life longer than that of the first member and an activity greater than that of any other member including the first at any time during transportation; in that case, the transport group of the nuclide "x" and the activity of the mixture shall be the maximum activity of that nuclide "x" during transportation.

§ 173.391 Small quantities of radioactive materials and radioactive devices. (a) Radioactive materials in normal form not exceeding 0.01 millicurie of Group I radionuclides; 0.1 millicurie of Group II radionuclides; 1 millicurie of Groups III, IV, V, or VI radionuclides; 25 curies of Group VII radionuclides; tritium oxide in aqueous solution with a concentration not exceeding 0.5 millicuries per milliliter and with a total activity per package of not more than 3 curies; or 1 millicurie of radioactive material in special form; and not containing more than 15 grams of uranium-235 are exempt from specification packaging, marking, and labeling, and are exempt from the provisions of § 173.393, if the following conditions are met:

(1) The materials are packaged in strong tight packages such that there will be no leakage of radioactive materials under conditions normally incident to transportation.

(2) The package must be such that the radiation dose rate at any point on the external surface of the package does not exceed 0.5 millirem per hour.

(3) There must be no significant removable radioactive surface contamination on the exterior of the package (see § 173.397).

(4) The outside of the inner container must bear the marking "Radioactive."

(b) Manufactured articles such as instruments, clocks, electronic tubes or apparatus, or other similar devices, having radioactive materials (other than liquids) in a nondispersible form as a component part, are exempt from specification packaging, marking, and labeling, and are exempt from the provisions of § 173.393, if the following conditions are met:

NOTE 1: For radioactive gases, the requirement for the radioactive material to be in a nondispersible form does not apply.

(1) Radioactive materials are securely contained within the devices, or are securely packaged in strong, tight packages, so that there will be no leakage of radioactive materials under conditions normally incident to transportation.

(2) The radiation dose rate at four inches from any unpackaged device does not exceed 10 millirem per hour.

(3) The radiation dose rate at any point on the external surface of the outside container does not exceed 0.5 millirem per hour. However, for carload or truckload lots only, the radiation at the external surface of the package or the item may exceed 0.5 millirem per hour, but must not exceed 2 millirem per hour.

(4) There must be no significant removable radioactive surface contamination on the exterior of the package (see § 173.397).

(5) The total radioactivity content of a package containing radioactive devices must not exceed the quantities shown in the following table:

Transport group	Quantity in curies	
	Per device	Per package
I	0.0001	0.001
II	0.001	0.03
III	0.01	3
IV	0.05	3
V or VI	1	1
VII	25	200
Special form	0.003	20

(6) No package may contain more than 15 grams of fissile material.

(c) Manufactured articles, other than reactor fuel elements, in which the sole radioactive material is natural or depleted uranium, are exempt from specification packaging, marking, and labeling

and are exempt from the provisions of § 173.393, if the following conditions are met:

- (1) The radiation dose rate at any point on the external surface of the outside container does not exceed 0.5 millirem per hour;
- (2) There must be no detectable radioactive surface contamination on the exterior of the package (see § 173.397).
- (3) The total radioactivity content of each article must not exceed 3 curies.
- (4) The outer surface of the uranium is enclosed in an inactive metallic sheet.

NOTE: Such articles may be packagings for the transportation of radioactive materials.

(d) Shipments made under this section for transportation by motor carriers are exempt from Part 177, except § 177.817.

**§ 173.392 Low specific activity materials.** (a) Low specific activity materials, when transported on transport vehicles other than those assigned for the sole use of the consignor, are exempt from the provisions of § 173.393 (a) through (2); however, they must be packaged in accordance with the requirements of §§ 173.395, and must be marked and labeled as required in §§ 173.401 and 173.402.

(b) Low specific activity materials which are transported in transport vehicles (except aircraft) assigned for the sole use of that consignor are exempt from specification packaging, marking, and labeling provided the shipments meet the requirements of paragraph (c) or (d) of this section.

(c) Packaged shipments of low specific activity materials transported in transport vehicles (except aircraft) assigned for the sole use of that consignor must comply with the following:

(1) Materials must be packaged in strong, tight packages so that there will be no leakage of radioactive material under conditions normally incident to transportation.

(2) Packages must not have any significant removable surface contamination (see § 173.397).

(3) External radiation levels must comply with § 173.393 (j).

(4) Shipments must be loaded by consignor and unloaded by consignee from the transport vehicle in which originally loaded.

(5) There must be no loose radioactive material in the car or vehicle.

(6) Shipment must be braced so as to prevent leakage or shift of lading under conditions normally incident to transportation.

(7) Except for shipments of uranium or thorium ores, unconcentrated, the transport vehicle must be placarded with the placards prescribed in accordance with § 174.541 (b) or § 177.823 as appropriate.

(8) The outside of each outside package must be stencilled or otherwise marked "Radioactive—LSA."

(d) Unpackaged (bulk) shipments of low specific activity materials transported in closed transport vehicles (except aircraft) assigned for the sole use of that consignor must comply with the following:

(1) Authorized materials are limited to the following:

(i) Uranium or thorium ores and physical or chemical concentrates of those ores.

(ii) Uranium metal or natural thorium metal, or alloys of these materials; or

(iii) Materials of low radioactive concentration, if the average estimated radioactivity concentration does not exceed 0.001 millicurie per gram and the contribution from Group I material does not exceed one percent of the total radioactivity.

(iv) Objects of nonradioactive material externally contaminated with radioactive material, if the radioactive material is not readily dispersible and the surface contamination, when averaged over one square meter, does not exceed 0.0001 millicurie per square centimeter of Group I radionuclides or 0.001 millicurie per square centimeter of other radionuclides. Such objects must be suitably wrapped or enclosed.

(2) Bulk liquids must be transported in the following:

(i) Spec. 103C-W (§§ 179.200, 179.201, and 179.202) tank cars. Bottom fittings and valves are not authorized.

(ii) Spec. MC 310, MC 311, MC 312, or MC 331 (§ 178.330, § 178.331, § 178.337, or § 178.343) cargo tanks. Authorized only where the radioactivity concentration does not exceed 10 percent of the specified low specific activity levels (see § 173.393(c)). The requirements of § 173.393(g) do not apply to these cargo tanks. Bottom fittings and valves are not authorized. Trailer-on-flat-car service is not authorized.

(3) External radiation levels must comply with subparagraphs (2), (3), and (4) of § 173.393(j).

(4) Shipments must be loaded by the consignor, and unloaded by the consignee from the transport vehicles in which originally loaded.

(5) Except for shipments of uranium or thorium ores, unconcentrated, the transport vehicle must be placarded with the placards prescribed in accordance with § 174.541 (b) or § 177.823, as appropriate.

(6) There must be no leakage of radioactive materials from the vehicle.

**173.393 General packaging requirements.** (a) Unless otherwise specified, all shipments of radioactive materials must meet all requirements of this section, and must be packaged as prescribed in §§ 173.391 through 173.396.

(b) The outside of each package must incorporate a feature such as a seal, which is not readily breakable and which, while intact, will be evidence that the package has not been illicitly opened.

(c) The smallest outside dimension of any package must be 4 inches or greater.

(d) Radioactive materials must be packaged in packagings which have been designed to maintain shielding efficiency and leak tightness, so that, under conditions normally incident to transportation, there will be no release of radioactive material. If necessary, additional suitable inside packaging must be used. Each package must be capable of meeting the standards in § 173.395 (b) (see also § 173.24). Specification containers listed as authorized for radioactive materials shipments may be assumed to meet those standards, provided the packages do not exceed the gross weight limits prescribed for those containers in Part 178.

(1) Internal bracing or cushioning, where used, must be adequate to assure that, under the conditions normally incident to transportation, the distance from the inner container or radioactive material to the outside wall of the package remains within the limits for which the package design was based, and the radiation dose rate external to the package does not exceed the transport index number shown on the label. Inner shield closures must be positively secured to prevent loss of the contents.

(e) The packaging must be so designed, constructed, and loaded that, when transporting large quantities of radioactive material:

(1) The heat generated within the package because of the radioactive materials present will not, at any time during transportation, affect the efficiency of the package under the conditions normally incident to transportation, and

(2) The temperature of the accessible external surfaces of the package will not exceed 122° F. in the shade when fully loaded, assuming still air at ambient temperature. If the package is transported in a transport vehicle consigned for the sole use of the consignor, the maximum accessible external surface temperature shall be 180° F.

(f) Pyrophoric materials, in addition to the packaging prescribed in this subpart, must also meet the packaging requirements of § 173.134 or § 173.154. Pyrophoric radioactive liquids may not be shipped by air.

(g) Liquid radioactive material must be packaged in or within a leak-resistant and corrosion-resistant inner container. In addition—

(1) The packaging must be adequate to prevent loss or dispersal of the radioactive contents from the inner container, if the package were subject to the 30-foot drop test prescribed in § 173.398 (c) (2) (i); or

(2) Enough absorbent material must be provided to absorb at least twice the volume of the radioactive liquid contents. The absorbent material may be located outside the radiation shield only if it can be shown that if the radioactive liquid contents were taken up by the absorbent material the resultant dose rate at the surface of the package would not exceed 1,000 millirem per hour

(h) There must be no significant removable radioactive surface contamination on the exterior of the package (see § 173.397).

(i) Except for shipments described in paragraph (j) of this section, all radioactive materials must be packaged in suitable packaging (shielded, if necessary) so that at any time during the normal conditions incident to transportation the radiation dose rate does not exceed 200 millirem per hour at any point on the external surface of the package, and the transport index does not exceed 10.

(j) Packages for which the radiation dose rate exceeds the limits specified in paragraph (i) of this section, but does not exceed at any time during transportation any of the limits specified in subparagraphs (1) through (4) of this paragraph, may be transported in a transport vehicle (except aircraft) assigned for the sole use of that consignor, and unloaded by the consignee from the transport vehicle in which originally loaded.

(1) 1,000 millirem per hour at 3 feet from the external surface of the package (closed transport vehicle only);

(2) 200 millirem per hour at any point on the external surface of the car or vehicle (closed transport vehicle only);

(3) 10 millirem per hour at 6 feet from the external surface of the car or vehicle; and

(4) 2 millirem per hour in any normally occupied position in the car or vehicle, except that this provision does not apply to private motor carriers.

(k) When radioactive materials are loaded by the shipper into a transport vehicle assigned for the sole use of that shipper, the shipper must observe all applicable requirements of Part 174, 175, or 177, as appropriate.

(l) Packages consigned for export are also subject to the regulations of the foreign governments involved in the shipment. See §§ 173.8 and 173.9.

**§ 173.394 Radioactive material in special form.** (a) Type A quantities of special form radioactive materials must be packaged as follows:

(1) Spec. 5B, 5D, 6A, 6B, 6C, 6J, 6K, 6L, 6M, 17C, 17H, 42B, or 42C (§§ 178.82, 178.84, 178.97, 178.98, 178.99, 178.100, 178.101, 178.103, 178.104, 178.107, 178.108, 178.115, and 178.118) metal drums.

(2) Spec. 21C (§ 178.224) fiber drums.

(3) Spec. 14, 15A, 15B, 15C, 15D, 19A, or 19B (§§ 178.168, 178.169, 178.170, 178.171, 178.190, and 178.191) wooden boxes.

(4) Any Spec. 12 series (§§ 178.205 through 178.212) fiberboard boxes, 200-pound test minimum, or Spec. 23F or 23H (§ 178.214 or § 178.219) fiberboard boxes.

(5) Spec. 55 (§ 178.250) metal-encased shielded container. Additionally authorized for not more than 300 curies per package, for domestic shipments only.

(6) Spec. 7A (§ 178.350) Type A general package.

(7) Foreign-made packagings which bear the symbol "Type A" may be used for transportation of radioactive materials from the point of entry in the United States to their destination in the United States or through the United States en route to a point of destination outside of the United States.

(b) Type B quantities of special form radioactive materials must be packaged as follows:

(1) Spec. 55 (§ 178.250) metal-encased shielded container. Authorized only for not more than 300 curies per package. Authorized for domestic shipments only (see also § 173.394 (a) (5)).

(2) Spec. 6M (§ 178.304) metal packaging.

(3) Any Type B packaging specifically approved for such use by the Department.

(c) Large quantities of radioactive materials in special form must be packaged as follows:

(1) Spec. 6M (§ 178.304) metal packaging. Radioactive thermal decay energy must not exceed 10 watts.

(2) Any Type B packaging which meets the standards in the regulations of the U. S. Atomic Energy Commission (Title 10, Code of Federal Regulations, Part 71), or the 1967 regulations of the International Atomic Energy Agency, and which has been

specifically authorized for such use by the Department under Part 170. In applying for Departmental authorization of packages for large quantities of radioactive materials to be used in shipments by the U. S. Atomic Energy Commission, or one of its contractors or licensees, a copy of the license amendment or other approval issued by that Commission will be accepted in place of the package structural integrity evaluation.

**§ 173.395 Radioactive material in normal form.** (a) Type A quantities of normal form radioactive materials must be packaged as follows:

(1) Spec. 5B, 5D, 6A, 6B, 6C, 6J, 6K, 6L, 6M, 17C, 17H, 42B, or 42C (§§ 178.82, 178.84, 178.97, 178.98, 178.99, 178.100, 178.101, 178.103, 178.104, 178.107, 178.108, 178.115, and 178.118) metal drums.

(2) Spec. 21C (§ 178.224) fiber drums.

(3) Spec. 14, 15A, 15B, 15C, 15D, 19A, or 19B (§§ 178.165, 178.168, 178.169, 178.170, 178.171, 178.190, and 178.191) wooden boxes.

(4) Any Spec. 12 series (§§ 178.205 through 178.212) fiberboard boxes, 200-pound test minimum; or Spec. 23F or 23H (§ 178.214 or § 178.219) fiberboard boxes.

(5) Any Spec. 3 or 4 series (§§ 178.36 through 178.44 or §§ 178.47 through 178.58) cylinders.

(6) Spec. 55 (§ 178.250) metal-encased shielded container.

(7) Spec. 7A (§ 178.350) Type A general package.

(8) Foreign-made packagings which bear the symbol "Type A" may be used for transportation of radioactive materials from their point of entry in the United States to their destination in the United States or through the United States en route to a point of destination outside of the United States.

(b) Type B quantities of radioactive materials in normal form must be packaged as follows:

(1) Spec. 6M (§ 178.304) metal packaging. Authorized only for solid or gaseous radioactive materials which will not decompose at temperatures up to 250° F.

(2) Any Type B packaging specifically approved for such use by the Department.

(c) Large quantities of radioactive materials in normal form must be packaged as follows:

(1) Spec. 6M (§ 178.304) metal packaging. Authorized only for solid or gaseous radioactive materials which will not decompose at temperatures up to 250° F. Radioactive thermal decay energy must not exceed 10 watts.

(2) Any Type B packaging which meets the standards prescribed in the regulations of the U. S. Atomic Energy Commission (Title 10, Code of Federal Regulations, Part 71) or the 1967 regulations of the International Atomic Energy Agency, and which has been specifically authorized for such use by the Department under Part 170. In applying for Departmental authorization of package for large quantities of radioactive materials to be used in shipments by the U. S. Atomic Energy Commission, or one of its contractors or licensees, a copy of the license amendment or other approval issued by that Commission will be accepted in place of the package structural integrity evaluation.